Reactors and Nuclear Safety Unit



Aerial view of the Portuguese Research Reactor in early 1961.



From left to right: Jaime da Costa Oliveira, Júlio Pistacchini Galvão, Frederico Gama Carvalho, António Gonçalves Ramalho and Cândido Marciano da Silva, during the ceremony of the 50th anniversary of the Portuguese Research Reactor.

Reactors and Nuclear Safety Unit

José Gonçalves Marques

The year 2011 marks the 50th anniversary of the *Portuguese Research Reactor* (RPI). The reactor achieved its first criticality in the early hours of April 25, 1961, just two days before the inauguration of the then Laboratory for Nuclear Physics and Engineering. A small ceremony was held on April 27, 2011, with interventions of some of the key researchers in the early days and of the Minister for Science and Higher Education.

The Research Unit on Reactors and Nuclear Safety houses not only the RPI, but also the *Neutron Activation in Environment, Nutrition and Epidemiology* and *Applied Dynamics* groups. The RPI also supports activities for groups in the other Research Units of ITN, as well as in Universities.

A total of 6 researchers were hired under the *Ciência* initiative in 2008 and 2009, effectively doubling the number of full-time researchers in this Unit. However, these researchers reached 50% or more of their contracts during 2011.

The staff involved in all aspects of the operation and use of the RPI presents its activities under the common headline of *Operation and Exploitation of the Reactor*.

The *Neutron Activation in Environment, Nutrition* and *Epidemiology* group uses the k_0 INAA technique in the RPI and was again the main Portuguese user of the reactor in 2011, accounting more than 50% of the total irradiation time. The group is dedicated to

cycling and impact of trace elements in the atmosphere. It addresses, specifically, the development and application of nuclear techniques, source apportionment and tracking in the atmosphere, chemical speciation, uptake and release of chemical elements in biomonitoring and monitoring, as well as health linkage through epidemiology and nutrition studies. These objectives are approached through research, included mostly in Ph.D. theses. The activities are essentially financed by the Foundation for Science and Technology (FCT).

The research performed by the Applied Dynamics group is mostly concerned by vibration and acoustic problems displayed by components of nuclear and conventional power plants. As such, a significant part of their research results has been motivated and funded by the French Commissariat à l'Energie Atomique (CEA) and the Portuguese Electricidade de Portugal (EDP). However, the techniques developed by this group can and have been used to solve problems, both of industrial and fundamental nature, outside the realm of power generation, e.g., the development of advanced methods for tuning and restoration of the Mafra carillons. In spite of continuing to be one of the smallest groups in terms of ITN staff, this fact is compensated by an active collaboration with Universities and Research Laboratories, both in Portugal and abroad. The vitality of this group is well demonstrated by their research contracts and publications.

Staff

Researchers

A.G. MARQUES, Princ
A. FALCÃO, Princ.
A. KLING, Princ. (85%)
A.V. ANTUNES, Princ.
M.C. FREITAS, Princ. (retired in 2011)
N.P. BARRADAS, Princ. (85%)
A.C. FERNANDES, Aux.
A.R. RAMOS WAHL, Aux. (85%)
D. BEASLEY, Aux.
H.M. DUNG, Aux.
M.A. S. PEREIRA, Aux.
S.M. ALMEIDA, Aux.
V. DEBUT, Aux.

Admin. & Techn. Personnel

A. RODRIGUES F.B. GOMES I. DIONÍSIO J.A. M. RIBEIRO J.C. ROXO J.P. SANTOS N. SERROTE R. POMBO R. SANTOS T. FERNANDES

OPERATION AND EXPLOITATION OF THE REACTOR

José Gonçalves Marques

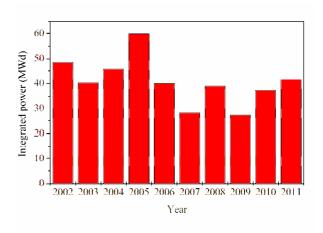
The main objective of the Operation and Exploitation of the Portuguese Research Reactor Group is to be able to satisfy the users' needs while conducting all tasks with the assurance that the reactor is operated in a safe and reliable manner by a highly competent and motivated staff. The implementation of such objectives demands a variety of activities, some of which are repetitive in objective and variable in content, while others address specific aspects of the same end situation.

User	Area	Time (%)
URSN	NAA	51.0
	Tomography	19.0
	Radiation effects	10.5
	Dosimetry and detector	5.3
	development	
	Education and training	1.2
UCQR	NAA	6.6
U. Lisboa	Isotope Production	0.5
	Detector development	0.5
IVIA	Radiation Effects	0.2
UPV	Isotope Production	1.6
U. Heidelberg	Dating	3.6

The main users of the reactor are described in the Table above. The largest sustained activity supported by the RPI is neutron activation analysis (NAA) in the URSN and UCQR Research Units of ITN. Most other activities suffer large fluctuations.

Education and training is very dependent on the number of students that attend courses using the reactor in practical sessions. This activity had a significant boost in 2011 from the ICARO intensive course organized at ITN for 21 European students.

Activities using neutron beams are currently restricted to neutron tomography and irradiation of electronic components and systems. These two activities accounted for 30-40% of the reactor utilization in the last two years. A setup for Prompt Gamma Neutron Activation Analysis is being optimized. It is expected that this setup will contribute significantly to new activities in the RPI.



The integrated power in 2011 increased again, as shown in the figure above, marking this year as the best since 2005. The total irradiation time in 2011 was 1518 hours, corresponding to an average of 1.5 irradiations performed simultaneously during the normal working hours of the RPI.

Research Team

Researchers

J.G. MARQUES, Princ. A. FALCÃO, Princ. N.P. BARRADAS, Princ. (85%) A. KLING, Aux. (85%) A.P. FERNANDES, Aux A.R. WAHL, Aux. (85%) D. BEASLEY, Aux. M. PEREIRA, Aux.

Reactor Operators

J.A.M. RIBEIRO J.C. ROXO N. SERROTE R. SANTOS

Technical Personnel

A. RODRIGUES F.B. GOMES J.P. SANTOS R. POMBO

Students

M.A.F. COSTA, Ph.D. Student UNL, FCT grant

Collaborators

T. GIRARD, CFNUL

Neutron tomography activities at the RPI

M.A. Stanojev Pereira, J.G. Marques

Objectives

The neutron radiography (NR) and Neutron tomography (NT) are very efficient tools to inspect the internal structure of materials. An equipment for neutron tomography was installed in the RPI under project POCI/FIS/59287/2004, funded by FCT, in the horizontal access of the thermal column and provides a parallel beam having 5 cm in diameter at the sample irradiation position. Since 2010 this equipment is operational and for the present conditions, the irradiation time to obtain an image is 90 seconds and the spatial resolution is about 300 μm.

Results

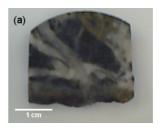
Imaging techniques can be classified according to the penetrating radiation type, such as X - ray, gamma - ray, and neutron and because of their attenuation characteristics, in many cases, they are complementary methods.

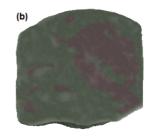
Neutrons are strongly attenuated by H-rich substances even when wrapped by thick metal layers while for X rays the inverse occurs. Hence neutrons are commonly used to inspect, e.g., orings, components of refrigerators, fuel cells, aeronautical devices and cultural heritage objects. For neutron tomography it is necessary to obtain about 200 single 2D images over 180 degrees. In our setup these images are captured using a Peltiercooled CCD camera.

The figure 1 shows an example of a 3D image obtained at the RPI. This object is a tile treated with the consolidant Paraloid B-72 and shows clearly the penetration of the resin. The main conclusions of this study are published¹ and also discussed in a contribution from the Chemical Radiopharmaceutical Sciences Unit.

The technique also allows to inspect mineral veins. The figure 2 shows preliminary data for a copper ore, from the Department of Mineralogy of the Faculty of Science at the University of Lisbon. The figure 2a is a photography and 2b and 2c are tomography images of the metal, highlighted in red.

neutron tomography images and after resin treatment.





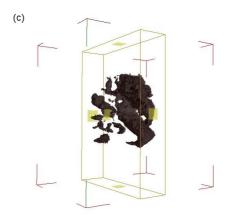


Fig 2. (a) Photography of a Cu ore; (b) tomographic image 1 mm from surface; (c) detailed distribution of Cu.

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⁽a) (c) Gray - surface Red - ceramic body Blue - voids 1 cm Green - resin Fig. 1. Fragment of glazed tile (16th century) from Museu Nacional do Azulejo: (a) untreated; frontal (b) and angular (c)

Prudêncio, M.I., Stanojev Pereira, M. A., Marques, J.G., Dias, M.I., Esteves, L. Burbidge, C.I., Trindade, M.J., Albuquerque, M.B.. Neutron tomography for the assessment of consolidant impregnation efficiency in Portuguese glazed tiles (16th and 18th centuries). Journal of Archaeological Science (2011), doi:10.1016/j.jas.2011.11.010.

Fast neutron irradiation of GaN HEMT transistors

Q. Vinckier¹, J.G. Marques, C. Cruz

Gallium nitride (GaN) High Electron Mobility Transistors (HEMT) are fast becoming adopted for high power amplifier applications. The key advantage of these transistors is their operating power density, which is significantly higher than the one of Si-based transistors. However, not much is known about the behaviour under radiation of GaN-based transistors, which only became commercially available in recent years. A methodology was developed to simulate the defects introduced by neutrons using the MCNPX and SRIM Monte Carlo codes, linked by a custom-made FORTRAN program. HEMT GaN transistors from two manufacturers (Cree and EPC) were irradiated with fast neutrons in the RPI, with fluences up to $2x10^{14}$ n/cm² (E> 1 MeV). Si based MOSFET transistors were irradiated at the same time for comparison. Characteristic curves were obtained during irradiation using a custom-made test platform, able to supply the required high drain current. The results show significantly different behaviours for the two HEMT transistors based on similar materials and general geometrical arrangements. Detailed results were presented in the M.Sc. thesis of Q. Vinckier at ISIB, Brussels.

¹M.Sc. student from ISIB, Belgium

Intensive course on accelerator and reactor operation (ICARO)

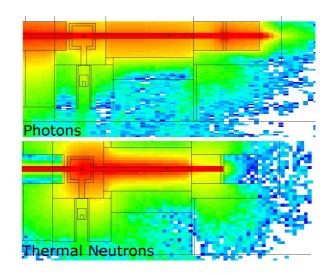
J.G. Marques, E. Alves, P. Vaz

ITN hosted for the second time the "Intensive Course on Accelerator and Reactor Operation" (ICARO), from 6 to 19 March 2011 for the CHERNE (Cooperation for Higher Education on Radiological and Nuclear Engineering, http://www.upv.es/cherne/) network, with financial support of the Erasmus Intensive Program. Twenty four students from the Applied University of Aachen, University of Bologna, University of Catania, University of Coimbra, University of Palermo, University of Prague, University of Valencia, ISIB (Institut Supérieur Industriel de Bruxelles), POLIMI (Politecnico di Milano), and XIOS (eXpertisecentrum voor Industrie, Onderwijs en Samenleving) attended the course in 2011. The course included 16 theoretical classes and 34 practical classes on radiation protection, accelerator operation and applications, as well as research reactor operation and applications. Two round tables were also organized on the topics "Nuclear as Part of Sustainable Development?" and "Ethics and the Principle of Justification: The Case of Nuclear Technology Assessment".

Modelling of a proposed beam line extension for multi-elemental Prompt Gamma Neutron Activation Analysis

D.G. Beasley, A.C. Fernandes, J.G. Marques, A.R. Ramos Wahl, J. Santos

Prompt Gamma Neutron Activation Analysis (PGNAA) is seen as a technique with added value to the activities based on the RPI. However, the background at the Ge detector position is too high, preventing the practical use of this technique. A proposal to extend the beam_line and build a new detector port and sample chamber was simulated. Grids of foils (Au, Ni, Al and In) were placed in the existing installations to verify and normalize simulations performed using MCNPX. TLDs were placed inside the chamber to measure the photon dose. The simulations allowed for the model to be optimized, the shielding requirements to be determined and for a significant drop in background at the new detector position. Expected flux at the detector positions were calculated from simulations to assist in planning the experimental parameters. The work is expected to be completed in early 2012.



Implementation of a radiological surveillance scheme for the supervised areas at URSN

A. Kling, A. R. Ramos, J. G. Marques

While an extended monitoring for the radiation levels in the controlled area of URSN (the reactor hall) has been operational for many years, a systematic surveillance scheme for the four supervised areas (the annex for radioactive effluent tanks and three laboratories in the main building) was lacking. Based on information provided by the Radiation Protection Technicians (effluent tanks) and the laboratory users for each area, suitable points have been selected and marked for the control of the gamma radiation level and the surface contamination. The surveillance is performed weekly in the early afternoon of Monday. This corresponds to the time period at which, due to the established work schedules, the largest number of radioactive samples and the maximum cumulated activity is present at the laboratories. The procedures and maps containing the designated measuring points have been included in the Radiation Protection Plan of URSN. The measured data are collected on a dedicated form, evaluated by the Responsible for the Radiological Protection of URSN and reported in an adequate form in the annual radiological control report.

Study on the reactor production of 163Ho needed in the search for neutrinoless double Beta-decay A. Kling, M.R. Gomes¹, F. Gatti²

In the neutrinoless double beta decay two neutrons decay simultaneously within a nucleus into two protons and two electrons – a process that is forbidden in the Standard model due the law of lepton number conservation. The proof of the existence of such a decay scheme and consequently the neutrino being a Majorana particle (i.e. particle and antiparticle are identical) is therefore of highest interest. Various approaches with suitable isotopes (e.g. 3 H, 76 Ge, 130 Te, 187 Re) and detection techniques (e.g. electrostatic spectrometers, enriched HPGe detectors, cryogenic calorimeters) have been made so far. A very promising candidate is 163 Ho due to its low Q-value (2.80 keV) and relatively short half-life (4570 y) to be used with a cryogenic detector developed by a group in Genoa. A reactor-based production of this isotope is possible through the 162 Er (n, γ) 163 Er reaction with subsequent β -decay to 163 Ho. However, it is found that the low natural isotopic abundance of 162 Er (0.14%) makes it impossible to use natural Er as target. On the other hand, calculations showed that with commercially available targets enriched to 40% in 162 Er specific activities of about 2 kBq/mg, Er can be achieved at RPI after 120 hours of irradiation. Furthermore the radiocontamination in this case will be basically restricted to 165 Er which decays with a half-life of ca. 10 hours to stable 165 Ho and therefore does not impose any limitations in the decay experiments.

The SIMPLE dark matter search project

M. Felizardo, T.A. Girard¹, A.C. Fernandes, A. Kling, A.R. Ramos, J.G. Marques, M. Auguste², D. Boyer², A. Cavaillou², C. Sudre², J. Poupeney², H.S. Miley³, R.F. Payne³, J. Puibasset⁴, F.P. Carvalho, M.I. Prudêncio, R. Marques

SIMPLE project activity in 2011 centered on the analysis of the previous Stage 2 dark matter search data from the underground site (Laboratoire Souterrain Bas Bruit, LSBB). New radioassays of the various shielding materials were completed and incorporated into the previous MCNP neutron background estimate. A re-analysis of the data obtained at the RPI yielded an improved detector nucleation efficiency. The Stage 2 results were reported at the 2011 TAUP meeting in Munich, Germany, and submitted to Phys. Rev. Lett.; combined with those of Stage 1, the results provided the current most stringent limits against a spin-dependent WIMP coupling (overlapping for the first time with limits obtained by indirect means), and eliminated a large part of the recent CoGeNT claim for a light mass WIMP discovery. New single-detector neutron and α calibration measurements were performed, confirming the results of the 2010 Phys. Rev. Lett. obtained with separate detectors. Publicized critiques by J. Collar and by E. Dahl et. al (COUPP Collaboration) were publicly responded to. Measurements of the unshielded neutron background in the GESA site of the LSBB were initiated by the team of V. LaCoste (IRSN) in November, using a set of 13 Bonner spheres.

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¹Centro de Física Nuclear, Universidade de Lisboa, 1649--003 Lisbon, Portugal

² Laboratoire Souterrain á Bas Bruit (Universite de Nice-Sophia Antipolis), 84400 Rustrel, France

³ Pacific Northwest National Laboratory, Richland, WA 99352 USA

³ CRMD-CNRS, Université d' Orleans, 45071 Orleans, France

Applied Dynamics

José Antunes

The activities at Applied Dynamics Laboratory (ADL) are devoted to research in nuclear engineering, with an emphasis on the vibratory and acoustic behaviour of mechanical components. Our group started in 1986, with the following objectives: (1) Develop theoretical methods, computer tools and experimental techniques, to solve structural problems in nuclear power station components; (2) Use this state-of-the-art know-how, in order to solve structural problems arising in Portuguese power plants and other industrial facilities.

The first objective has been pursued through extensive international collaboration with our main scientific partner - the French Commissariat à l'Energie Atomique (CEA) / Département de Mécanique et Technologie (DMT). More than one decade of fruitful collaboration is attested by a significant number of published results. Important problems have been solved, such as nonlinear vibrations in steamgenerators, flow-induced vibrations of nuclear fuel and stability problems in rotating machinery. Furthermore, new identification techniques have been developed and applied with success to nonlinear dynamical systems.

The second objective has been pursued by starting in 1990 a series of projects with (and for) the Portuguese power supplier Electricidade de Portugal / Companhia de Portuguesa de Produção Electricidade (EDP/CPPE), stemming from actual structural problems in power plants (Sines, Setúbal).: These projects enabled us to model and solve vibratory problems arising in rotating machinery, vibroacoustical problems in boilers and heat-exchangers, as well as structural identification problems. Several computer codes have been developed in connection with these projects.

In recent years we also developed research projects of more fundamental nature, mainly funded through the Portuguese Science Foundation (FCT) research programmes. These projects have been developed in partnership with several Portuguese institutions (Faculdade de Ciências de Lisboa, Instituto Politécnico do Porto, Instituto Politécnico de Setúbal, Instituto Superior Técnico, Universidade Nova de Lisboa), as well as the Université de Paris, Trinity College Dublin and Southampton University. This

work, developed in the context of fundamental physics – in particular addressing problems in music acoustics, optimization and structural geology – is centred in modelling nonlinear dynamics and flow-structure phenomena. The methods developed transcend the context of these projects and may be adapted to solve several aspects of industrial problems.

The Applied Dynamics team is mainly concerned with the following scientific fields: structural dynamics, flow-induced vibrations, nonlinear dynamics, vibroacoustics, experimental methods, signal processing, system identification, structural and acoustical optimization. As a spin-off from our research activities, teaching has been actively pursued on structural dynamics and acoustics - ranging from university level courses in Portugal (Coimbra, Lisbon) to several post-graduation short courses abroad (Paris, Dublin, Cargèse). Also, student and post-doc training, as well as several university thesis (M.Sc. and Ph.D.) have been successfully supervised, for both Portuguese and foreign students. An extensive book on fluid-structure dynamics and acoustics, co-authored by two researchers from CEA and ITN/ADL was internationally published in 2006 and another volume on flow-induced vibrations is currently under completion, to be released in 2012.

Among the above-mentioned scientific fields one should stress those features which give this small group a distinct profile from others working in structural dynamics in Portugal. Those features are: (1) a proven expertise and output in flow-excited systems and nonlinear vibrations; (2) a complementary theoretical/experimental approach for every problem.

Most of the research projects pursued at ADL have been based on both industry and academic research contracts. Research activities at ADL were internationally recognized by two prizes from the American Association of Mechanical Engineers (ASME).

A new researcher, Vincent Debut, joined the permanent staff of the Applied Dynamics group in 2008 under a five year contract, being involved since then in most of the research activities at ADL.

Research Team

Researchers

J. ANTUNES, Princ., Group Leader V. DEBUT, Ass.

Students

M. CARVALHO, FCT grant

Collaborators

L. HENRIQUE, IPS, Adj. Professor M. MOREIRA, IPS, Adj. Professor O. INÁCIO, IPP., Adj. Professor M. MARQUES, IST, FCT grant

Simulation and experimental identification of flow-excited vibro-impact dynamics in nuclear components with support clearances

J. Antunes, V. Debut, P. Piteau¹, X. Delaune¹, L. Borsoi¹

Objectives

In nuclear facilities, flow turbulence excitations and fluid-elastic phenomena may induce structural vibrations or instabilities, leading to fatigue failures and wear, which must be addressed with particular care for safety reasons. At ITN/ADL, in a long-term close collaboration with CEA-Saclay (contract TGV-ICE), we gained significant expertise in these areas. As in previous years, this project was aimed at the development at ITN/LDA of up-to-date computational and identification techniques to deal with turbulenceinduced and fluid-elastically unstable vibrations of nuclear components, such as fuel rods or steamgenerator tubes. Effective use of our remote identification techniques was made in connection with validating experiments performed by colleagues at the test-loops in CEA-Saclay, France.

Results

The experimental setup sketched and pictured in Figure 1 was used as a representative conceptual rig for steam-generator tubes. A flexible instrumented beam was excited by a shaker or by air flow, leading to vibro-impact responses. Figure 2 shows a detail of the instrumented clearance support.

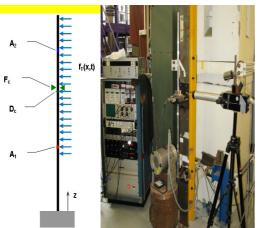


Fig. 1 - Test rig developed at CEA-Saclay for experimental validation of the impact force and response identification

techniques developed at ITN/ADI



Fig. 2 - Detail of the instrumented clearance support of the experimental rig at CEA-Saclay.

Figure 3 illustrates one such experiment, showing the impulsive impact forces and the gap-limited tube vibrating motion, for a turbulence excited test. The experimental and remotely identified results obtained constitute a validation of our identification approach to deal with gap-supported tubes subjected to flow excitation, at least for a single clearance support. We are currently extending this work to multi-supported systems.

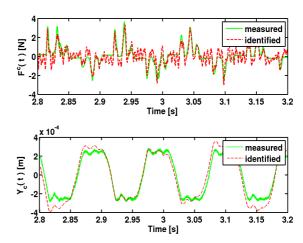


Fig. 3 - Measured and remotely identified impact force and tube displacement at the gap-support, under turbulence excitation of the vibrating beam.

Results connected with this research were published at several international conferences and journal papers, as next listed.

Publications

- X. Delaune, P. Piteau, V. Debut, J. Antunes, "Experimental validation of inverse techniques for the remote identification of impact forces in gapsupported systems subjected to local and flow turbulence excitations", ASME Journal of Pressure Vessel Technology, Vol. 5, Paper 051301 (2011).
- J. Antunes, X. Delaune, P. Piteau, "Time-domain simulation of the random vibrations of tubes subjected turbulence-conveying flows", Paper 57162, Proceedings of the ASME 2011 Pressure Vessels and Piping Conference (PVP 2011), July 17-21, 2011, Baltimore, USA.
- X. Delaune, J. Antunes, P. Piteau, L. Borsoi, "Experiments and computations of a loosely supported tube under two-phase flow buffeting and fluid-elastic coupling forces", 21st International Conference on Structural Mechanics in Reactor Technology (SMIRT 21), 6-11 November 2011, New Delhi, India.
- P. Piteau, X. Delaune, J. Antunes, L. Borsoi, "Experiments and computations of a loosely supported tube in a rigid bundle subjected to singlephase flow", Jounal of Fluids and Structures (in print).

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¹ Commissariat à l'Energie Atomique, DEN/DM2S/SEMT/DYN, CEN-Saclay, France

Dynamical analysis and tuning of carillon bells

J. Antunes, V. Debut, M. Carvalho, O. Inácio¹, J. Soeiro de Carvalho²

In the context of the FCT project PTDC/EAT-MMU/104255/2008, we have started a three-year project dealing on the physical and musicological aspects of the historical Mafra carillons, from the XVII century. After development of a specific modal identification software, well adapted to axi-symmetrical shells, which was validated by laboratory experiments performed on a contemporary bell, we started the identification field-work in the Mafra carillons, which will be pursued during 2012 in parallel with the development of tuning techniques.



Fig. 1 - Experimental identification of bell modes: laboratory bell (left), Mafra carillon (right).

Analytical computation of shell modes subjected to fluid-structure coupling

J. Antunes, X. Delaune¹, P. Piteau¹

Fluid-structure coupling problems are recurrent in nuclear power facilities (steam-generators, pipes, reactor vessels, pressurizing systems). In the context of a research project funded by CEA/Saclay (France) we have developed a theoretical model to predict the modes of cylindrical shells when subjected to coupling by an incompressible dense fluid. This theoretical model was validated against extensive finite-element computations using the CAST3M program, and then applied in the analysis of vibratory experiments performed at Saclay.

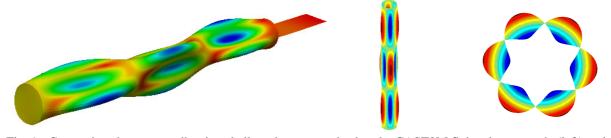
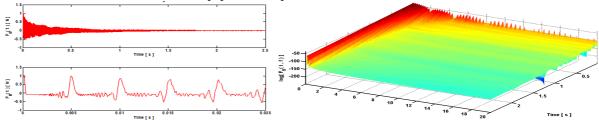


Fig. 1 - Comparison between a vibration shell mode computed using the CAST3M finite element code (left) and the corresponding results from the simplified analytical model (right).

Dynamics and string/body coupling in the Portuguese guitar

J. Antunes, V. Debut, O. Inácio¹, M. Marques², A. Ribeiro², P. Serrão²

In the context of the FCT project PTDC/FIS/103306/2008, we started in 2011 a detailed analysis of the dynamics of worn strings, which display local mass-deposits and/or mass-voids at specific locations due to playing. We developed a detailed model for worn strings, capable of computing their distorted vibration modes and simulating their time-domain responses. A detailed statistical analysis of the dynamical changes due to the string wear was also achieved. A conference paper will be presented soon on this work.



Time-domain bridge excitation force from a plucked string (left) and the corresponding spectrogram (right).

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Instituto Politécnico do Porto, Escola Superior de Música e Artes do Espectáculo, Laboratory of Music Acoustics, Porto

² Universidade Nova de Lisboa, Faculdade de Ciências Sociais e Humanas, Lisbon

¹ Commissariat à l'Energie Atomique, Laboratory of Dynamical Studies, Saclay, France

Instituto Politécnico do Porto, Laboratory of Music Acoustics, Porto; ²Instituto Superior Técnico, Lisbon

Neutron Activation in Environment, Nutrition and Epidemiology

Marta Almeida

The research in NANE group is focused on the development of the NAA (Neutron Activation Analysis) methodologies and on their application to environment, nutrition and epidemiology studies. NANE is a multidisciplinary group that has the collaboration of researchers from different fields of science: environment, chemistry, environmental health, biology, biochemistry and geology. The capability of NAA techniques at RPI is based on irradiation and measurement facilities that allow the utilization of the entire neutron spectrum of the reactor, epithermal neutrons (ENAA), cyclic (CNAA), Compton irradiations suppression spectroscopy (CSS), automatic sampling changers (ASCs) and prompt gamma (PGAA) (ongoing work). Nowadays, the combination of NAA techniques in ITN permits the determination of the elements Ag, Al, As, Ba, Br, Ca, Cd, Ce, Cl, Co, Cr, Cs, Cu, Dy, Eu, F, Fe, Hf, Hg, I, K, La, Lu, Mn, Na, Nd, Rb, Sb, Sc, Se, Sm, Sr, Ta, Tb, Ti, U, V, Yb, Zn and Zr in biological, environmental and material samples.

The unit activities include the following scientific interests:

Quality assurance and quality control (QA/QC), development of methodologies and automatization.

The principal objective of the group is to assure the quality of the results given by the NAA techniques. Within the activities of the NANE group, a large number of researchers/students is involved, principally doing temporary studies. These researchers deal with a big number of samples and with several equipments and softwares in order to generate large databases. For this reason, the QA/QC is essential to guarantee the quality of the generated data. To accomplish this objective Inter and Intra- laboratory exercises are performed in a routine basis. This line also aims to optimize the analytical techniques *in* order to improve the analysis of the samples, to speed the calculations and to simplify the handling of the data.

Environment

Over the last two decades, NANE group have developed aerosol sampling and analysis

instrumentation, which nowadays are applied successfully for measurements of various physical and chemical aerosol parameters. NANE research have already shown that NAA can be advantageously used in aerosol studies because many elements are determined, high accuracy and precision are obtained, many samples are analyzed and little sample preparation is necessary.

At the moment, the complete characterization of the particles and biomonitores at the receptor is advantageously used by NANE group to elucidate the sources of the pollutants and the processes associated with their formation, to assess local, regional and long-range transport and, finally, to identify mitigation options focusing on the reduction of the air pollutant concentrations.

Epidemiological studies

The objective of this research line is to establish unequivocal associations between pollution, morbidity and mortality. Respiratory problems, cardiovascular disease and carcinogenic incidence in the Portuguese population have been studied in association with chemical elements measured by NAA.

Nutrition

Selenium has been the target of NANE research in the nutrition field because it is an essential element to humans and, in Portugal, its contents in diet are deficient. NANE group is studying different types of Selenium supplementation and is evaluating their efficiency.

Training

The research unit has a strong component in graduation and post-graduation training (B.Sc., M.Sc., Ph.D., post-Ph.D.).

Services

NANE provide analytical services under request for private national and international companies and universities. In these services NAA characterize materials and environmental samples.

Research Team

Researchers

S.M. ALMEIDA, Aux. (Contract), Group Leader H.M. DUNG, Aux. (Contract)

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Selenium distribution in cereals and portuguese cultivation soils. Interactions between selenium and iodine uptake by cereals - a case study.

C. Galinha¹, M.C. Freitas, A.M.G. Pacheco¹, J. Coutinho², B. Maçãs², A.S. Almeida²

Objectives

Selenium is a trace element essential to the well-being and health quality of humankind. This project focuses on determining the ability of bread (*Triticum aestivum* L.) and durum wheat (*Triticum durum* Desf.) to accumulate selenium after supplementation.



Fig. 1 - Outline of mainland Portugal, showing the approximate location of the experimental fields.

Two of the most representative wheat cultivars in the country -- Jordão (bread) and Marialva (durum) -- have been selected for field trials, following the same agronomic practices and schedules as the regular (non-supplemented) crops of those varieties. Soil supplements were applied at the sowing time, whereas foliar additions were performed at the booting and grain-filling stages, in either case by using sodium selenate and sodium selenite solutions at three different Se concentrations, equivalent to field supplementation rates of 4, 20 and 100 g of Se per ha (with and without potassium iodide, as a joint additive). All field experiments were done in south areas of mainland Portugal (Fig. 1).

Results

Seed enrichment may be viewed as an alternative to classical biofortification strategies, such as soil amendments or foliar treatments. A study has dealt with the first phase of such an alternative, that is, with the preparation of Se-enriched seeds of wheat by optimizing both their soaking time (in an active Se solution) and washing time (in bidistilled water), through detection of a Se radiotracer (75Se). The study has been designed to conform to realistic Sesupplementation rates and to have an extension in actual field trials. The optimized times for administering Se to seeds (soaking time) and ensuring inner-seed levels only (washing time) are 48 and 24 h, respectively.

After harvesting, Se contents in these cereal grains were compared to data from regular wheat samples (field blanks) grown at the same soil/time, yet devoid of any supplements. Total Se in all field samples -- a three-fold replication, in a full-factorial design -- have been determined by instrumental neutron activation analysis, via the short-lived nuclide 77m Se (half-life time: 17.5 s).

The results show that foliar additions can increase Se contents in mature grains up to 35 times, when compared to non-supplemented crops, and that selenate-based treatments seem more effective in enhancing such contents (Fig. 2 and 3). Jordão and Marialva may respond differently to the stage of application though.

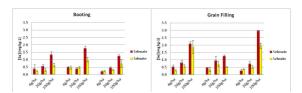


Fig. 2 Selenium results for foliar supplementation in Marialva

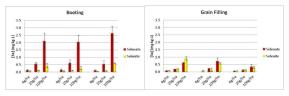


Fig. 3 Selenium results for foliar supplementation in Jordão.

Overall, for any given field supplementation rate, foliar application appears to translate into higher Se concentrations in wheat grains than each soil counterpart.

Published work

C. Galinha, M.C. Freitas, A.M.G. Pacheco, J. Kameník, J. Kucera, H.M. Anawar, J. Coutinho, B. Maçãs, A.S. Almeida (2011) Selenium in cereal plants and cultivation soils by radiochemical neutron activation analysis. Journal of Radioanalytical and Nuclear Chemistry, doi: 10.1007/s10967-011-1262-0

- C. Galinha, M.C. Freitas, A.M.G. Pacheco, J. Coutinho, B. Maçãs, A.S. Almeida Radiotracing selenium in bread-wheat seeds for a Sebiofortification program: an optimization study in seed enrichment. Journal of Radioanalytical and Nuclear Chemistry, 291(2011) 193-195.
- C. Galinha, M.C. Freitas, A.M.G. Pacheco, J. Coutinho, B. Maçãs, A.S. Almeida, Determination of selenium in bread-wheat samples grown under a Sesupplementation regime in actual field conditions. Journal of Radioanalytical and Nuclear Chemistry, 291(2011) 231-235.
- C. Galinha, H.M. Anawar, M.C. Freitas, A.M.G. Pacheco, M. Almeida-Silva, J. Coutinho, B. Maçãs, A.S. Almeida, Neutron activation analysis of wheat samples, Applied Radiation and Isotopes, 69 (2011) 1596-1604.

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¹ CERENA-IST; ² INRB/INIA-Elvas

Development of k_0 -based cyclic NAA for short-lived radionuclides

H.M. Dung, S.M. Almeida, I. Dionisio, D.G. Beasley, M.C. Freitas, J. Marques

The k_0 -based cyclic neutron activation analysis (k_0 -CNAA) technique has been studied at the Portuguese research reactor (RPI) for the determination of elements which form short-lived radionuclides, particularly fluorine (20 F, 11.16 s half-life) and selenium (77m Se, 17.36 s half-life) in polymer, biological and environmental samples. The detection limits obtained for F and Se were about 50 and 0.01 mg kg⁻¹, respectively, in the investigated materials. The timing parameters for the procedure were 10 to 20 s for irradiation, 5 s decay, 10 to 20 s counting, and 5 s waiting and performed with 8 cycles. The k_0 -IAEA program was modified to use millisecond time resolution for irradiation, decay and counting times as needed for interpreting k_0 -CNAA data in terms of concentration, accuracy and detection limit. The analytical quality of the procedure was evaluated by the analysis of standard reference materials with the accuracy within 15% for F and Se and within 12% for other elements producing short-lived or detectable radionuclides, i.e. Al, Ca, Cl, Cu, Dy, I, Mg, Mn, Ti, and V.

Quality control of k_0 -NAA by participating in the inter-comparison study (WEPAL, the Netherlands) *H.M. Dung, S.M. Almeida, I. Dionisio, M.C. Freitas*

A total of 8 dried samples of plant (IPE) and soil (ISE) materials have been received via an IAEA proficiency test programme organised by WEPAL, the Netherlands. The samples were analysed with 6 replicates for each sample by the k_0 -NAA procedures developed at RPI. The average results of the determinations have been reported to the IAEA and the WEPAL after 6 weeks of the arrival of the samples. The results have been processed at Wageningen University and published under code names. Determinations were made for the following elements: Ag, Al, As, Ba, Br, Ca, Ce, Co, Cr, Cs, Cu, F, Fe, Ga, Hg, I, K, La, Mg, Mn, Mo, Na, Nb, Nd, Pd, Rb, Sb, Sc, Se, Sn, Sr, Th, Ti, U, V, W, Y, Zn and Zr.

Optimization of the Neutron Activation Analysis capabilities

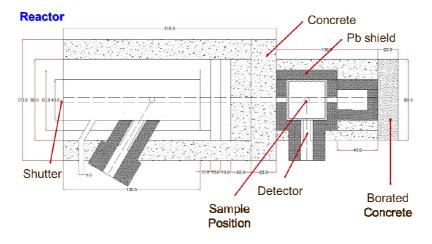
H.M. Dung, S.M. Almeida, M.C. Freitas

A new CANBERRA detector (GC-3018) was purchased and installed in one of the automatic sample changers (ASC2-N#2). The dead time of the gamma-ray spectrometers has been improved after re-design the ground system between DSpecs and the ASC2s. The spectrometers' electronic parameters have also been optimized in order to enhance the spectral quality and stability. The permanent database used for k_0 -IAEA software has regularly been updated in order to meet the requirements of the applications of interest.

Enhancing the sustainability of RR & their safe operation through regional cooperation, networking & coalitions, IAEA TC Regional Project RER/4/032

N. Canha, H.M. Dung, M.C. Freitas, S.M. Almeida, J. Marques, Zs. Révay¹

This project supported by the IAEA has the goal to implement a PGNAA (Prompt Gamma Neutron Activation Analysis) facility at the RPI-ITN. A new design of the facility was created to overcome several problems detected in preliminary studies (such as high background) and the necessary materials to build it were acquired. In October 2011, a visit from an IAEA team was conducted to ITN to discuss this project and its achievements and to define further timelines.



¹KFKI, Budapest, Hungary

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Impact of atmospheric indoor aerosol in human health, PTDC/SAU-ESA/65597/2006

N. Canha, M. Almeida-Silva, S.M. Almeida, M.C. Freitas, M. Martinho¹, P. Pegas², C. Alves², C. Pio², M. Trancoso³, R. Sousa³, F. Mouro³, T. Contreiras ⁴

A statistical treatment was conducted over the database of indoor air parameters and the primary schools where these parameters were sampled (urban area of Lisbon, Portugal). The aim of this study was to assess the associations between indoor air parameters with the schools building characteristics, through the use of statistical methods. Several associations were found and allow pointing out several recommendations to improve the classrooms IAQ. Overall, the conclusions of this study point to the following recommendations: 1) classrooms should face streets rather than patios and should not be located in basements, 2) the density of students can affect the indoor environment and should be limited, 3) wooden materials appear to have advantages as building materials due to a lower input of contaminants. Other factors, like ventilation, cleanings and use chalk versus whiteboard pens, can reduce or increase specific contaminants inside the classrooms.

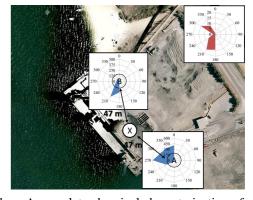
¹United Nations-NY/USA, ²UA/CESAM, ³LNEG/UB-LBA, ⁴INSA/ONSA

Measurement of fugitive emissions during harbour operations, PTDC/AAC-AMB/098825/2008

A.V. Silva, S.M. Almeida, A.M. Marques¹, A.I. Silva, C.A. Ramos¹, S. Almeida¹, A. Lopes¹, A.I. Pedro¹, A. Ferreira¹, T. Pinheiro, S. M. Garcia², G. Domingues², A.I. Miranda³

Fugitive emissions pose problems both for general air quality management and for the operational management of the facilities. In harbours, activities such as loading, unloading and transport of dusty materials are important sources of particles fugitive emissions.

The aim of this study was to estimate the impact of harbour activities on Air Particulate Matter (APM) levels and composition. This work was based on experimental campaigns carried out in a Portuguese harbour when three types of bulk materials – fertilizer, phosphorite from Syria and phosphorite from Morocco - were handled. High time resolution monitors were installed close to the unloaded area and recorded APM concentrations meteorological variables. PM_{2.5} and PM_{2.5-10} were also collected



during unloading operations, in polycarbonate filters by Gent samplers. A complete chemical characterization of collected samples was made by the techniques Instrumental Neutron Activation Analysis and Particle Induced X-Ray Emission.

Results showed that manipulation of materials during harbour operations resulted in high emissions of particles, especially from the coarse fraction. These emissions were very affected by the granulometry of the handled materials and by the meteorological conditions.

Escola Superior de Tecnologia da Saúde de Lisboa ²Instituto de Soldadura e Qualidade ³CESAM, Universidade de Aveiro.

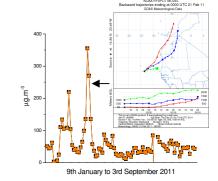
CV-Dust - Atmospheric aerosol in Cape Verde region: seasonal evaluation of composition, sources and transport, PTDC/AAC-CLI/100331/2008

M. Almeida-Silva, S.M. Almeida, T. Sitoe, M.C. Freitas, C. A. Pio¹, T. Fernandes¹, D. Custódio¹, J. Cardoso²

Cape Verde is located in an area of massive dust transport from land to ocean. Thus, it is ideal to set up sampling devices that will allow the characterization and the quantification of the dust transported from Africa. To

characterize the chemical composition of dust transported from Africa, a total of 100 Nucleopore and Teflon filters were sampled in Praia, Cape Verde. All filters were weighted by gravimetry in a controlled clean room (class 10,000) with a semi-micro balance. Filter mass before and after sampling was obtained as the average of three measurements, when observed variations were less than 1%.

Hysplit Model, ending in Praia in 3rd February 2011 confirms an air mass transported from Sahara Desert localized in the Center of Africa. Other higher peak of particles concentrations happed in 21st February 2011 and the Hysplit Model confirms an air mass transported from Sahara Desert but also from Atlas Mountains located in the North of Africa.



²Cape Verde University

¹CESAM – Aveiro University

Integration of biomonitoring and instrumental techniques to assess the air quality in an industrial area located in the coastal of central Asturias, Spain, RFSR-CT-2009-00029

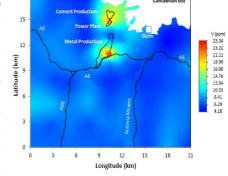
S.M. Almeida M.C. Freitas, A.I. Pedro, T. Ribeiro, J. Lage, A. V. Silva, N. Canha, M. Almeida-Silva, T. Sitoe, I. Dionisio, S. Garcia¹, G. Domingues¹, J. Perim de Faria², B. González Fernández², D. Ciaparra³

A biomonitoring study was performed within the European Project "Assessment of Emissions and Impact of Steel Processes" in order to indicate geographical variances in trace-element air pollution around the Arcelor Mittal steelwork placed in Gijon, Spain.

Lichens were transplanted from a clean background site, exposed in an industrial area in Gijon, and collected after 5 months. After lichens exposure, the study had three objectives: 1) to explore spatial patterns of lichens conductivity in order to identify the impact of the industrial studied area; 2) to study the spatial distribution of chemical elements determined by INAA technique; 3) to establish a relationship between the distribution of conductivity and the concentrations measured in lichens and the characteristics of local sources.

Results showed that the values of conductivity and antropogenic

elements were significantly higher near the main industries (a steel plant, a power plant and a cement plant).

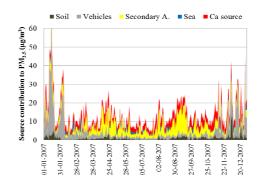


¹ISQ, ²Arcelor Mittal Spain, ³Tata Steel,

Characterizing seasonal variations in elemental particulate matter concentrations in european urban and rural areas under different climatic conditions, IAEA RER/2/005

S.M. Almeida, N. Canha, M.C. Freitas, I. Dionisio, T. Sitoe, J. Cardoso¹, A. Caseiro¹, C.A. Pio¹, H.Th. Wolerbeek²

The aim of the project is to apply non-destructive and multielemental nuclear techniques to characterize APM in different countries in Central and Western Europe, to contrast the results for areas with a continental and a marine climate and to compare them with the acceptable standards for particulate matter and elemental concentrations as defined by European countries. The meteorological parameters have also been measured and correlated with the element concentrations. In Portugal, APM was collected in 1 rural (Foros do Arão) and 2 urban (Lisbon and Setúbal) areas and element concentrations were measured by Neutron Activation Analysis in ITN. APM and element concentrations showed a seasonality characterized by higher



contributions of secondary aerosol during summer/spring and higher contributions of vehicles during autumn/winter. Several approaches were used in order to identify the emission sources: receptor models, air mass trajectory analysis; weekday/weekend ratios; hourly variations and comparison between stations.

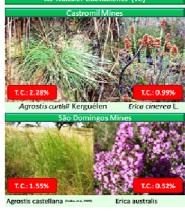
¹CESAM/Aveiro University; ²Dept. Radiation, Radionuclides & Reactors, Delft, The Netherlands

$(Hyper) accumulation \ of \ arsenic \ and \ other \ elements \ in \ plants \ adapted \ to \ sites \ impacted \ by \ mining \ and \ smelting \ activities \ (HYPERAS), PTDC/AMB/65462/2006$

N. Canha, H.M. Anawar, M.C. Freitas, S.M. Almeida, AI. Dionísio, H.M. Dung, .M.G. Pacheco¹, C.J. Pinto-Gomes², A. Bettencourt³

This study aimed to find out a vascular plant species that accumulate relatively high concentrations of arsenic (As) for its use as phytoremediator at abandoned and contaminated mining areas, such as São Domingos mines (1), Portugal. The assessment of As contamination levels in soils and plants of other similar sites in the north of the country (Castromil-2 and Poço de Freitas-3) was also conducted; and the sample analyses were made by instrumental neutron activation analysis. *Agrostis* genera have shown higher As transfer coefficients than other studied plant species and, in particular, *Agrostis curtisii* has shown a reasonable ability to accumulate high concentration of this toxic element.

¹CERENA/IST, ²ICAM - Évora University, ³CMA-IMAR, Évora Univ



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