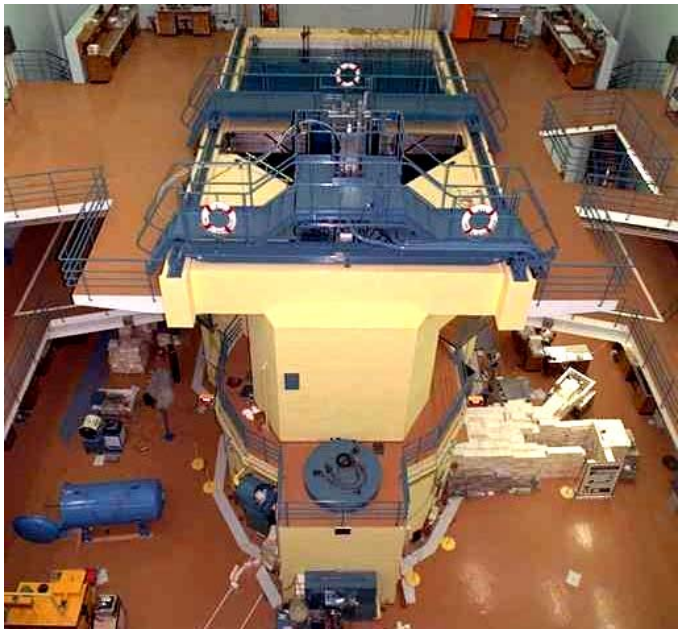
Reactor Sector



Reactor

José Carvalho Soares

The Portuguese Research Reactor (RPI), as a unique infrastructure in the Iberian Peninsula, houses as well the ***Atmospheric Elemental Dispersion*** and ***Applied Dynamics*** groups. The RPI also supports activities in the Chemistry and Physics Sectors. Foreign users from Universities and Research Laboratories accounted for 21% of the total irradiation time of the RPI, which was the largest increase in the reactor utilisation.

The Reactor was the first sector to have an infusion of personnel that was essential not only to keep the RPI running, but also to reinforce its research activities. Another young researcher was hired in 2001 and will start in January 2002. Two Principal Researchers have also been appointed this year and will start soon. The four new reactor operators trained with grants from the PRAXIS XXI program were hired, together with four new technicians for the operation and maintenance of the RPI. A new operators course is under final approval, meeting the challenge of personnel renewal.

Dr. António Ramalho has continued to coordinate the team which presents its activities under the headline of ***Operation and Exploitation of the Reactor, Dosimetry (RPI) and Reactor Calculations***. The number of research projects has doubled in 2001, corresponding to a threefold increase in funding, clearly benefitting from the presence of young researchers.

The ***Atmospheric Elemental Dispersion*** group is the main user of the RPI, accounting for 37% of the total irradiation time performed in the reactor. This group also makes an important bridge with the Physics Sector, complementing the NAA work done in the Reactor with PIXE in the van de Graaff accelerator. The group has a large number of graduate students and performs several services by contract, from which we highlight the monitoring of the atmosphere around the waste incinerator of São João da Talha.

The ***Applied Dynamics*** group is one of the smallest groups in terms of ITN staff, since it has only one researcher and one technician. This is compensated with an active collaboration with Universities and Research Laboratories both in Portugal and abroad. The vitality of this group is well demonstrated in their list of research contracts.

Structure of the Sector and Technical staff

Research groups in the Reactor Sector

– Operation and exploitation of the Reactor, Dosimetry (RPI) and Reactor Calculations
– Applied Dynamics
– Atmospheric Elemental Dispersion

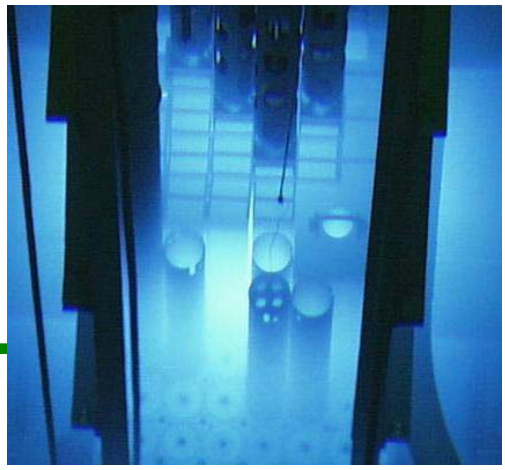
Other Research Staff

– Jaime Oliveira (Coordinator Researcher)
– Eduardo Martinho (Principal Researcher)

Administrative and Technical staff

– Odete Mendes
– Teresa Fernandes
– Albano Silva

Operation and Exploitation of the Reactor



Operation and Exploitation of the Reactor, Dosimetry (RPI) and Reactor Calculations

A.J.G. Ramalho

The central objective of the operation and exploitation of the reactor is to be able to satisfy the users needs while conducting all activities with the assurance that a highly competent and motivated staff operates the equipment in a safe and reliable manner. The implementation of such objectives demands a variety of projects, some of which are repetitive in objective and variable in content, while others address specific aspects of the same end situation. Safety, being a permanent consideration in all work around the reactor, interlinks all the activities being or to be performed.

The main set of projects, actual and coming, in which the staff is involved is presented below.

The programme for testing the behaviour of electronic components to be used at the LHC/CERN, simulating the expected exposure for a 10 years period, has continued during 2001 with the irradiations being performed in a beam tube that was prepared for the effect and put into operation last year.

An Emission Channeling/Blocking Setup for the investigation of the lattice of single crystals as well as intrinsic and extrinsic defects has been designed, and will commence installation soon.

Isotope production had a decreased importance in the reactor utilization. Effort continued to be placed in the production of short lived isotopes, which can be delivered rapidly to the users in the country and in Spain.

The operation of the reactor requires updated reactor physics calculation of parameters such as effective multiplication, reactivity margins, control rod worth, fuel consumption, and safety parameters. The main drive of the work was the preparatory studies relating to the core configuration change that will take place in early 2002.

There has been significant activity in the field of dosimetry and BNCT. The activity in dosimetry has also addressed the characterisation of the irradiation facilities, and obtained values for validation of calculations, to refer some examples. A PhD dissertation in this field is close to completion.

The implementation of an epithermal neutron beam has progressed considerably. The external concrete shielding was installed, as well as the in-pile collimators and filters. Several measurements were done to define the dimensions of the external collimator and of the protection of the active neutron detectors. The construction of the relevant pieces is underway.

Concerning the installation of neutron physics equipment, the double axis spectrometer (DIDE) is operational since last spring, and the in-pile components of the small angle difractometer (EPA) were recently installed.

The main users of the reactor are described in Table I. Fig. 1 indicates the reactor usage in terms of the number of hours of irradiations and the number of irradiations performed.

Table 1

User	Area	Time (%)
ITN-RPI	NAA	37.1
	Dosimetry and BNCT	13.2
	Other (training, etc)	4.4
ITN-Chemistry	Isotope Production	2.6
	NAA	11.4
ITN-Physics	Beam Users	1.3
	Materials testing	9.6
Univ. Coimbra	Isotope Production	<0.1
CERN-LHC	Circuits Irradiation	8.7
Univ. Manchester	Dating of rocks	7.3
Univ. Paris VII	Detectors calibration	4.3
Industry (Spain)	Isotope Production	<0.1

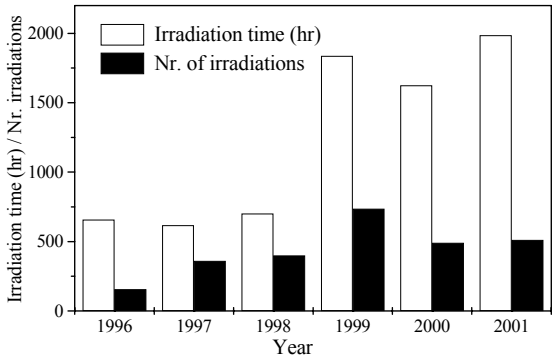


Fig. 1

In the last three years there has been a significant increase in these numbers. It is worth noting that, in 1999, the integrated power was 44 MWd, in 2000 it was 50 MWd, and in 2001 (until mid-December) it was 52 MWd.

It is also interesting to note a continued increase of users from abroad (Spain, France, Switzerland, UK), representing about 21% of the total time.

Operation and Exploitation of the Reactor, Dosimetry (RPI) and Reactor Calculations

Research Team

Researchers

- A.G. RAMALHO, Principal Researcher, Group leader
- I.C. GONÇALVES, Auxiliary Researcher
- F. CARDEIRA, Auxiliary Researcher
- J.G. MARQUES, Auxiliary Researcher
- A. KING , Auxiliary Researcher (90%)
- N.P. BARRADAS, Auxiliary Researcher (95%)

Collaborators

- C. CHAUSSY
- JEAN-M. CHAUSSY

Reactor Operators

- M.C. MARQUES
- R. CARVALHO
- J.A.M. RIBEIRO
- J.C. ROXO
- N. SERROTE
- V. PÁSCOA
- R. SANTOS

Visiting Researchers

- P. VAZ, Auxiliary Professor, IST, Lisbon
- A. VIEIRA, Co-ordinator Professor, ISEP, Porto

Students

- M.J. PRATA, PhD Student, IST, FCT grant
- A. FERNANDES, PhD Student, IST, FCT grant
- J.A. SANTOS, BSc Student, ITN, ITN grant
- R. PATRÍCIO, PhD Student, IST/ITN

Technical Personnel

- R. POMBO
- V. TOMÁS
- A. RODRIGUES (since October)
- J. S. SOUSA (since October)
- L.C. SANTOS (since October)
- J. L. SANTOS (since October)

Funding

	$\times 10^3$ PTE
Research Projects:	34357
ITN:	9240
Services:	1711
Total:	**45308**

Publications

Books:	2
Journals:	3 and 3 in press
Proceedings:	3
Conference communications:	4
Internal Reports:	1
Safety analyses:	2

Thermoluminescent dosimetry in mixed fields of neutrons and photons

A.C. Fernandes, I.C. Gonçalves, J.A. Santos, A.F. Carvalho, L. Santos, J. Cardoso

Objectives

Measurement of radiation doses at the RPI facilities in the experimental conditions.

Results

The study of the response of several TL materials in different irradiation conditions has been pursued. As an exemple, Figure 1 shows the reproducibility of GR100 detectors (^{nat}LiF:Mg,Ti) irradiated in two conditions:

a) pure gamma field;
b) alternate irradiations in a pure gamma field and in a mixed field of photons and neutrons.

The gamma field is the Co-60 source of LMIRR/ITN and the mixed field is the vertival access of the thermal column of RPI.

The recent acquisition of a TL reader with the possibility of glow-curve analysis will allow to obtain results at high dose level. This is the case for the high temperature peak in the glow curve of aluminium oxide detectors, as shown in Figure 2 for a dosemeter irradiated with 10 kGy of ^{60}Co.

An interesting goal for the future work is to study the response of TL materials in mixed fields of photons and neutrons with different energy spectra.

In this study the following irradiation facilities of RPI will be used:

a) vertical access of the thermal column as a nearly pure thermal neutron spectrum;

b) fast neutron beam at E4 irradiation tube, with a neutron spectrum where the intensity of the fast component is approximatly the double of the thermal and epithermal ones;

c) epithermal neutron beam at D1 irradiation tube where the thermal component may be cut with a cadmium filter, and the epithermal is the double of the fast one.

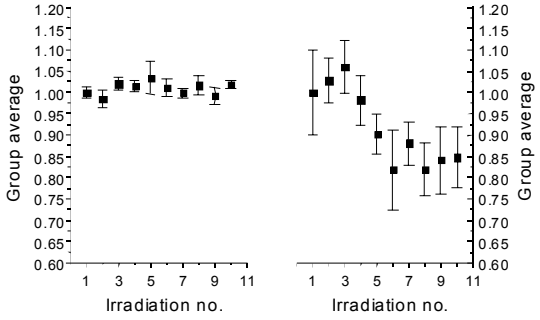


Fig. 1 – Reproducibility of GR-100 detectors over ten irradiations with 250 mGy of ^{60}Co. Left: consecutive photon irradiations. Right: alternated photon and mixed-field irradiations.

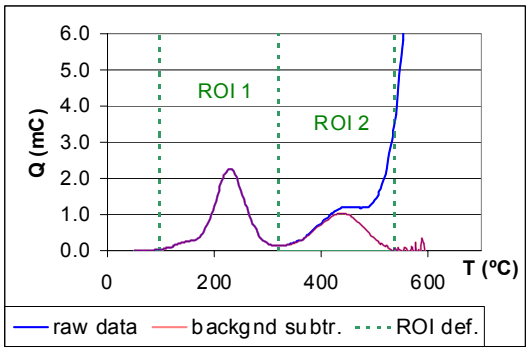


Fig. 2 – Glow curve of an aluminium oxide dosemeter irradiated with 10 kGy of ^{60}Co, showing the two dosimetric peaks.

Published, accepted or in press work

1. A.C. Fernandes, I.C. Gonçalves, A. Ferro de Carvalho, et al., Reproducibility of TL measurements in a mixed field of photons and neutrons, *XIII Int.Symp.on Solid State Dosimetry Athens 2001; Radiation Protection Dosimetry (in press).*

Monte Carlo modelling of the RPI core and validation with experimental results

A.C. Fernandes, I.C. Gonçalves, J.A. Santos

Objectives

Simulation of the reactor core with Monte Carlo code MCNP. Validation of the model with experimental results and comparison with deterministic calculations.

Results

The MCNP-4C code was used to simulate the RPI core. Criticality calculations were performed and compared with the results obtained with the deterministic code WIMS-D5 usually used in neutronic studies (Table 1). Two models of the fuel elements were used (Fig.1):

a) the regions with fissile material were homogenised;
b) the detailed description of the fuel plates was used

The code was used to calculate the neutron spectrum inside the fuel elements and at the grid positions.

These results are to be compared with those obtained experimentally with the multiple-foil activation method. In this method several detectors are used with sensitivities in the different energy intervals of the neutron spectrum (Table 2). Then the neutron spectrum is unfolded with the codes SANDII and LSL-M2. Experimental results were obtained with activation detectors irradiated in the spaces between the fuel plates and in the grid positions, along the height of the fuel elements. Figure 2 shows the calculated and measured response profiles for Mn and Au foils at fuel element N10.

The obtained model allows to calculate the neutron spectrum at the RPI core and to obtain the source term for subsequent simulations of the radiation fields at irradiation facilities of RPI.

Element	MCNP (homog.)	MCNP (detailed)	WIMS
Rod C1	0.0254 (3)	0.0252 (3)	0.02436
Rod C2	0.0256 (3)	0.0245 (3)	0.02466
Rod C3	0.0419 (3)	0.0411 (3)	0.03923
Al reflector	-	0.0028 (2)	0.0032

Table 1. Reactivity worth of control rods and aluminium reflector in cfg. N2-P1/6 of RPI.

Detector (dim. in mm)	*X-section (b)*	*Reaction and Response (MeV)*
Mn-Al 0.1% (0.1×dia.5)	σ_0=13.3 I_0=14.0	$^{55}Mn(n,\gamma)^{56}Mn$ 5.5E-9 - 3.40E-4
Au-Al 0.1% (0.1×dia.5)	σ_0=98.65 I_0=1550	$^{197}Au(n,\gamma)^{198}Au$ 1.5E-8 - 5.8E-6
In (0.127×dia.10)	σ_f=122E-3	$^{115}In(n,n')^{115m}In$ 1.0 - 5.6
Ni (0.05×dia.10)	σ_f=102.3E-3	$^{58}Ni(n,p)^{58}Co$ 1.9 - 7.5
Al (0.127×dia.10)	σ_f=0.685E-3	$^{27}Al(n,\alpha)^{24}Na$ 6.5 - 12

Table 2 - Activation detectors and reactions used for the dosimetry measurements at RPI. In the cross section column, I_0, σ_0 and σ_f are, respectively, the resonance integral, thermal and fission equivalent (in a ^{235}U fission spectrum) cross sections. 90% of response in an MTR spectrum is also shown.

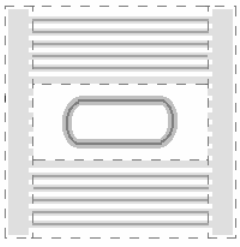


Fig 1. Cross sectional view of a control fuel element as simulated for MCNP. The homogenised regions are shown in dash.

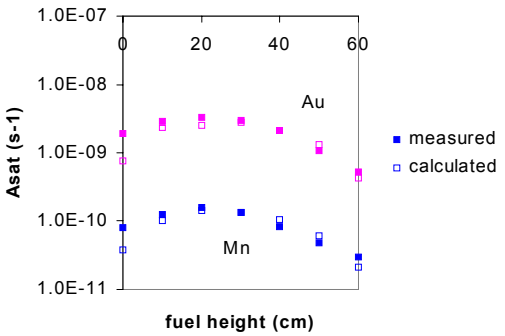


Fig 2. Measured and calculated response profile of Mn and Au foils irradiated at fuel element N10. Position of control rods=80%.

BNCT studies at RPI

I.C. Gonçalves, A.J.G. Ramalho, A.P. Fernandes, J.G. Marques, M.J. Prata, J.A. Santos, N. Oliveira[2], M. Castro[2], J. Rueff[1]

Objectives

The multidisciplinary BNCT project is being developed in collaboration with national and international teams. Implementation of dosimetric procedures and study of the effects in biological samples, due to the capture reaction in boron, are among the objectives of this project.

Results

The implementation of an epithermal neutron beam was already completed. It was installed in an irradiation tube of RPI and, according to Monte Carlo calculations, the intensity of the epithermal component is approximatly the double of the fast one. The optional introduction of a cadmium filter allows us to use the beam without thermal component.

The characterisation of the radiation field of this beam is being performed. Some preliminary results for the neutron fluxes are presented elsewhere ("Implementation of a multipurpose beam at RPI").

The twin ionisation chamber is the reference method in dosimetry of mixed fields. The optimisation of the conditions for the usage of a Mg/Ar and TE/TE ionisation chambers is being performed in agreement with the standards defined in an European Project for "Dosimetry for BNCT".

The study of genotoxic effects in human melanoma cells, due to the neutron capture reaction in boron, has been pursued. The vertical access of the thermal column of RPI is the irradiation facility used so far. This work is being conducted in collaboration with teams from Faculties of Pharmacy and Medicine of the Lisbon University and from the Faculty of Medical Sciences of the New University of Lisbon.

The future work includes:

a) to perform the complete characterisation of the radiation field of the epithermal beam;

b) to implement the TLD technique to measure radiation doses in free-beam and in phantom;

c) to pursue the work with biological samples, eventually by using epithermal neutrons.

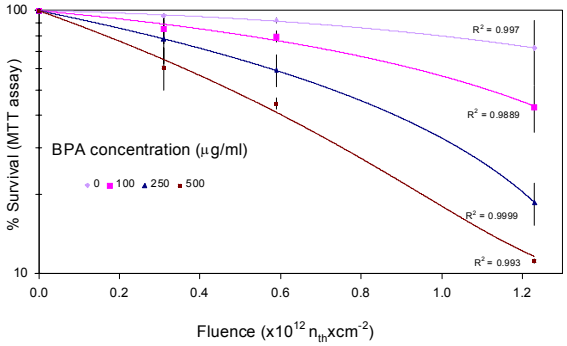


Fig.1 Cytotoxic effects on human melanoma cells induced by boron neutron capture reaction (thermal neutrons, vertical access of RPI)

Published, accepted or in press work

1. N.G. Oliveira, M. Castro, A.S. Rodrigues, I.C. Gonçalves, R. Cassapo, A.P. Fernandes et al., Evaluation of the genotoxic effects of the boron neutron capture reaction in human melanoma cells using the cytokinesis block. *Mutagenesis* vol.16 nº 5, pp 369-375, 2001.

2. N.G. Oliveira, M. Castro, A.S. Rodrigues, I.C. Gonçalves, R. Cassapo, A.P. Fernandes , Parp inhibitor 3-aminobenzamide does not increase the yield of chromosomal aberrant cells induced by boron neutron capture reaction inV79 chinese hamster cells. Presented at the 31th Annual Meeting of the European Environmental Mutagen Society. Ghent 2001. To be published in the revue of the Society.

3. N.G. Oliveira, M. Castro, A.S. Rodrigues, I.C. Gonçalves, On the mechanisms of genotoxicity of boron neutron capture reaction: data from human melanoma cells and non-tumoral cells. Oral presentation to the XXXII Reunião Anual da Sociedade Portuguesa de Farmacologia-XX Reunião de Farmacologia Clínica. Coimbra 2001.

[1]Dep.Genética, Fac.Ciências Médicas, Univ.Nova de Lisboa, Portugal
[2] Fac.Farmácia, Univ.de Lisboa

Implementation of a Multipurpose Beam Tube

J.M. Prata, A.J.G. Ramalho, J.G. Marques, A.P. Fernandes, I.C. Gonçalves

Objectives

Several applications require a neutron beam where the epithermal vs. fast component ratio is maximised. Among those applications is the determination of hydrogen at ppm levels in steels, by profiting of the anisotropic scattering of neutrons by hydrogen. Other envisaged applications are activities connected with Boron Neutron Capture Therapy and Boron determinations using Prompt Gamma Neutron Activation Analysis (PGNAA).

Results

The construction of the major mechanical components in the central workshop of ITN was finished only in May of this year and the whole setup was installed, although with a significant delay. The installed external shielding of the irradiation area was found to be appropriate, in good agreement with the Monte Carlo calculations performed.

The facility provides neutron beams with 2 and 5 cm diameter, with the possibility of leaving or eliminating the thermal component with a Cd filter. The irradiation area has dimensions of 40x40x125 cm^3. One of the lateral faces of the external shielding has an opening for an HPGe detector for PGNAA, placed at 60^o placed relatively to the incoming beam. Another opening in the external shielding allows the passage of cables such as those of the ^{3}He neutron detectors that will be used in the determination of H.

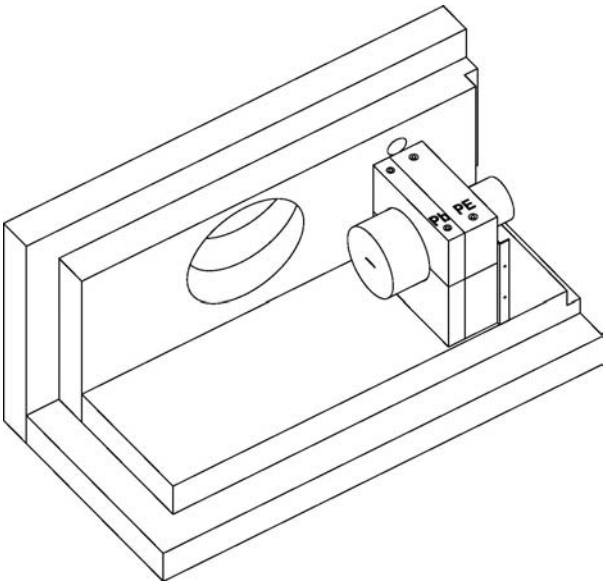


Fig. 1 View of the external collimator to be placed in the irradiation area.

The gamma dose rate was measured in several points in the irradiation area to define the dimensions of the gamma shielding for the ^{3}He detectors. The thermal and epithermal neutron fluxes were determined to be $3.6x10^7$ n/cm^2/s and $1.4x10^6$ n/cm^2/s, respectively, in reasonable agreement with the calculated values. For the determination of H in steels the neutron beam size will be reduced to 40 x 2 mm^2 using an external collimator, shown in Fig. 1, which is now under construction. The collimator and its base will also reduce the gamma field to less that 1 R/h in the area where the ^{3}He detectors will be placed. It is now antecipated that the first measurements of H concentration in steels will be done in the Spring of 2002.

Neutronics calculations for the RPI

N. P. Barradas, J.G. Marques, A.J.G. Ramalho

Objectives

The work developed along the year was centred in the study of situations required by the operational needs of the reactor. This involved primarily the following aspects: improve the neutron flux calculations, and the preparation of the core change to configuration N2-P1/7. We also studied the effect of Cd rods and voids in the vicinity of the core, as well as the absorbed dose in different materials.

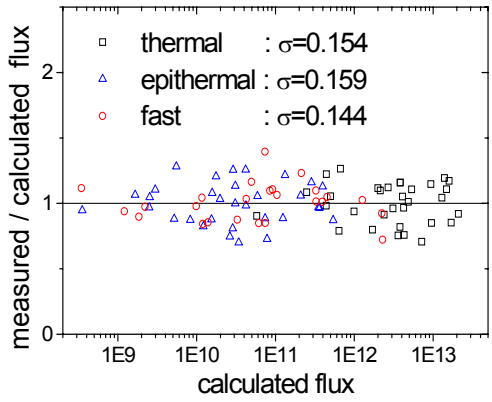


Fig.1. Dispersion of the ration between experimental and calculated flux values. The data span 11 years.

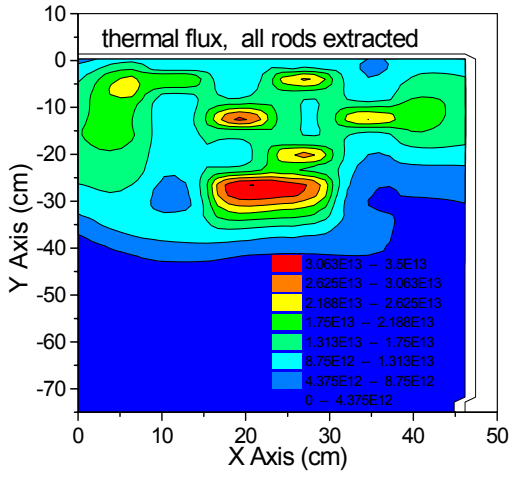


Fig.2. Calculated thermal flux for the proposed core configuration N2-P1/7

Results

There is a long standing issue of comparison of experimental and calculated thermal, epithermal and fast neutron fluxes in core 2 of the RPI, with a discrepancy between calculation and experiment, which has always been explained by a simple ad-hoc relation between the two sets of values. The procedure adopted until now has been to determine this relation using the few experimental points available for a given configuration, and then use it in all other grid positions to obtained "softened" calculated flux values. We have, on the one hand, reached a better agreement between calculation and experiment, and on the other hand, obtained a "universal" softening relation, able to explain all flux measurements so far made in core 2 of the RPI, which cover four configurations and over 10 years (see Fig. 1).

In the context of the core configuration change, around 125 configurations were analysed. The work involved [1]:

- Fuel replacement and reshuffling
- Moderator relocations
- Fuel burnup and configuration life time
- Replacement of the Al reflector by moderator and/or Be reflector to increase the expected usefull life of the configuration
- Flux calculations in the entire RPI grid
- Calculation of safety parameters

In particular, safety parameters were calculated for all intermediate configurations involved in the transition from configuration N2-P1/6 to N2-P1/7.

The "universal" softening relation was used to calculate the neutron fluxes in the proposed configuration N2-P1/7, shown in Fig. 2 for thermal neutrons.

The calculation of the absorbed dose of different materials in the mixed radiation field of the RPI was used in the safety report for irradiation of an experimental device containing Be [2].

Published, accepted or in press work

1. Coordinated by A.J.G. Ramalho; N.P. Barradas, F.M. Cardeira, J.G. Marques, A.J.G. Ramalho, *Alteração da configuração do núcleo do RPI - Configuração N2-P1/7 - Relatório de segurança, Internal Report,* Sacavém (2001).

2. M.R. da Silva, N.P. Barradas, A. Kling, J.G. Marques, A.J.G. Ramalho, *Irradiação de granalha de berílio: Análise de segurança, Internal Report,* Sacavém (2001).

Design of an Emission Channeling/Blocking Setup for the Portuguese Research Reactor

A. Kling, J.G. Marques, N.P. Barradas

Objectives

A setup for the investigation of the lattice of single crystals as well as intrinsic and extrinsic defects by emission channeling/blocking of charged particles (protons, tritons, alphas) from thermal neutron induced reactions at the Portuguese Research Reactor (RPI) facility has been designed. The method will enable high sensitivity studies of crystalline materials containing light elements like Li, B and N which are difficult to study by means of conventional ion beam techniques.

Results

Figure 1 shows a sketch of the emission channeling setup to be installed at the RPI. Since the expected count rates for charged particles generated in thermal neutron induced reactions are orders of magnitude lower than in a conventional Rutherford Backscattering experiment the choice of an appropriate detector is essential. A two-dimensional position sensitive detector with a large sensitive area (enables to place the detector at a sufficient distance from the sources of gamma and neutron radiation and enables the use of a 5×5 mm^2 beam spot) as well as a sufficient number of pixels (in order to obtain a good angular resolution) is therefore recommendable. In addition this detector type enables the recording of complete angular patterns that yield additional information on the lattice structure. The detector chosen for the setup is a Canberra doublesided strip detector with 3600 pixels on a 60×60 mm^2 area, which fulfills these requirements. At present the design for the readout electronics necessary for the recording of the angular patterns is going on.

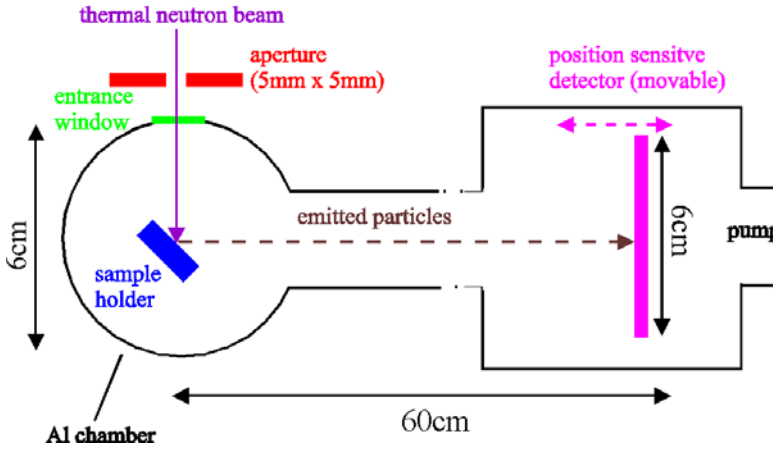


Fig. 1 Sketch of the experimental setup for emission channeling setup to be installed at the RPI.

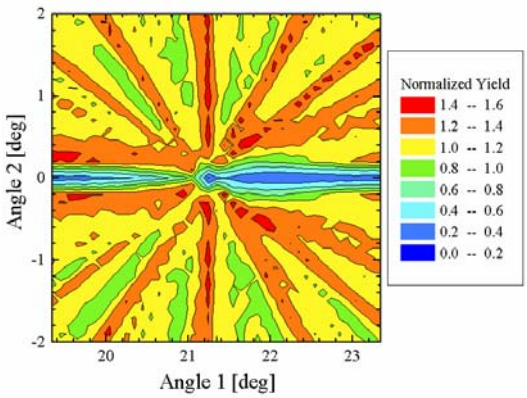


Fig.2 Calculated angular pattern for $LiNbO_3$.

In addition to the design of the experimental setup the simulation of the emitted charged particle angular distributions using Monte Carlo methods have been performed. Since emission channeling is the time-reversed effect of the conventional channeling effect it is possible to compute the patterns using the slightly modified code CASSIS that already exists at ITN. Figure 2 shows as an example the calculated angular pattern for the ^{6}Li(n,α)τ reaction in stoichiometric $LiNbO_3$ with the detector viewing the ⟨0441⟩ axis. Calculations on the influence of the intrinsic Li defect structure in the congruent type of this material are in progress.

Fast Neutron Irradiation of Electronic Circuits for the LHC/CERN

A.J.G. Ramalho, J.G. Marques, I.C. Gonçalves, A.P. Fernandes, N.P. Barradas, A. Vieira, M.J. Prata, F.M. Cardeira, J.A. Agapito[1], J.P. Santos[1], P. Gomes[2], J. Casas[2]

Objectives

Temperature measurement is a key issue in the LHC facility at CERN, as it will be used to regulate the cooling of the superconductor magnets. The signal conditioners for cryogenic thermometry are expected to receive a fast neutron fluence of the order of $2x10^{13}$ n/cm^2 during a 10 year period, as well as a gamma dose of 500 Gy, and this can affect the operation of the commercial circuits used in their construction. The operating conditions of these circuits are simulated using a fast neutron irradiation facility built in 2000.

Results

The fast neutron irradiation facility is installed in beam tube E4. The circuits are placed inside a cylindrical cavity, 100 cm long and 15 cm wide, inside the beam tube. The components under test were mounted on several PCBs, inside boxes. Each irradiation campaign ran from Monday to Friday with approximately 13 hours of irradiation followed by 11 hours of stand-by per day, due to the two-shift per day operation. The irradiation goal is to achieve a neutron fluence of $5x10^{13}$ n/cm^2 for the central PCB during one week of operation of the reactor with a gamma dose of 1.25 kGy to maintain the ratio estimated for the LHC. A forced-air cooling system was installed this year. The temperature during the irradiation of circuits decreased to about 40°C, which was crucial for the testing of analog-to-digital and digital-to-analog converters.

On-line measurements of properties of the circuits and components are performed before, during and after irradiation and stand-by periods, to evaluate the irradiation damages as well as possible annealing effects. The irradiation of components necessary for the construction of the signal conditioners continued in 2001 as foreseen. Several instrumentation amplifiers, voltage references, analog switches, and digital-to-analog converters were tested under irradiation [1].

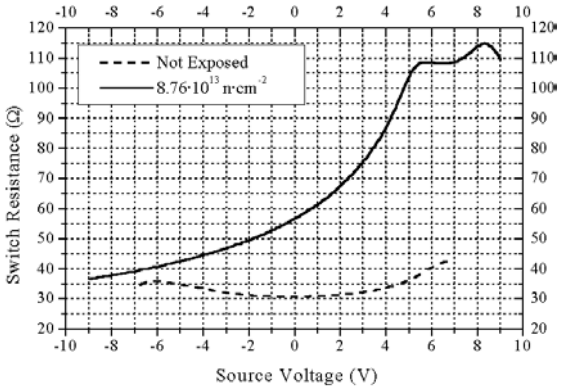


Fig 1. Analog switch resistance before and after irradiation.

Fig. 1 shows an example from an irradiated circuit. It was observed that the resistance of the switches of a DG412 device increased during the irradiation, rendering them useless. This behaviour could later be understood from the measurements shown in Fig. 1. The switch resistance vs. source voltage curve after irradiation is similar to that of a switch with a single operating MOS transistor (NMOS). Therefore it can be assumed that the threshold voltage of the PMOS transistor of the switch has been strongly modified and, consequently, it is always operating as an open circuit.

This work will continue with the testing of more components in statistically significant amounts. It is also foreseen that the testing of prototypes of some of the final circuit boards will start next year.

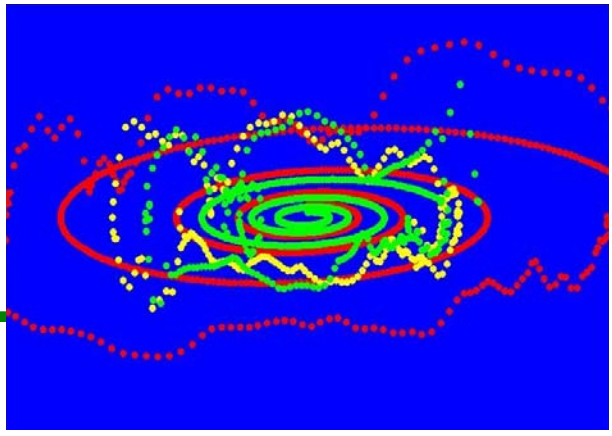
Published, accepted or in press work

1. J.A. Agapito, N.P. Barradas, F.M. Cardeira., J. Casas, A.P. Fernandes, F.J. Franco, P. Gomes, I.C. Gonçalves, A.H. Cachero, J. Lozano, J.G. Marques, A. Paz, M. J. Prata, A.J.G. Ramalho, M.A. Rodriguez Ruiz, J.P. Santos, A. Vieira, Radiation tests on commercial instrumentation amplifiers, analog switches and DACs, Proc. 7th Workshop on Electronics for LHC Experiments (http:// lebwshop.home.cern.ch/lebwshop/ LEB01_Book/LEB01_Proceedings.html).

[1] Universidad Complutense de Madrid

[2] CERN, LHC/ACR Division

Applied Dynamics



Applied Dynamics

José Antunes

The activities at "Applied Dynamics Laboratory" (ADL) are devoted to research in nuclear engineering, with an emphasis in the vibratory and acoustic behaviour of mechanical components. Our group started in 1986, with the following objectives:

- Develop theoretical methods, computer tools and experimental techniques, to solve structural problems in nuclear power station components;
- 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 steam-generators, 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 (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, vibro-acoustical 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 started several projects of more fundamental nature, in partnership with other Portuguese institutions, which have been funded through the Praxis XXI and Sapiens research programmes.

The Applied Dynamics team is mainly concerned with the following scientific fields: structural dynamics, flow-induced vibrations, nonlinear dynamics, vibro-acoustics, experimental methods, signal processing and system identification. As a spin-off from our research activities, teaching has been actively pursued on structural dynamics and acoustics — ranging from university level curses in Portugal (Coimbra, Lisbon) to several post-graduation short curses abroad (Paris, Dublin). Also, student training and university thesis (Graduation, MSc and PhD) have been successfully supervised, for both portuguese and foreign students.

Among the above-mentioned scientific fields one should stress those features which give our group a distinct profile from others working in structural dynamics in Portugal. Those features are:

- A proven expertise and output in flow-excited systems and nonlinear vibrations;
- A complementary theoretical/experimental approach for every problem.

There are no nuclear power stations in Portugal. However, past experience proved beyond doubt that the Applied Dynamics Laboratory is an active contributor at international science level, as well as a versatile problem-solving unit for domestic industrial partners. Therefore, the previously stated objectives remain our motivation and driving force. In spite of ADL extremely scarce permanent staff, we managed to attract motivated students and scientific collaborators from other institutions, which are essential for our activities. Beyond the close partnership with CEA, our work has been developed in collaboration with other Universities and Research laboratories, both in Portugal (IST, UNL, FCL, INESC, IPS, IPS, IPL, ENIDH) and abroad — France (ESPCI, Paris), Ireland (Trinity College, Dublin), England (Southampton University) and Greece (University of Tessalonika).

As in previous years, several research projects were started and pursued at ADL during 2001, all of them have been funded by research contracts:

- Project "Simulation and identification of complex dynamic systems (ROTDYN)" has been pursued, funded through the Sapiens programme;

- Project "Dynamical modelling of nonlinear systems (EXCITE)" has been pursued, funded through the Sapiens programme;

- Project "Dynamical modelling of geological inclusions (GEOMODELS)" was pursued, funded through the Sapiens programme;

- Project "Optimisation of rotor balancing" has been pursued, funded by EDP/CPPE;

- Project "Modelling of vibration-controlled encapsulation" has been pursued, funded by PROENOL;

- Project "Remote identification of impacts", formerly funded by CEA (France), was extended to investigate further aspects.

The average time-scale of ADL projects is 2~3 years. Several PhD thesis are being prepared in connection with them.

Applied Dynamics

Research Team

Researchers

- J.V. ANTUNES, Researcher, Group Leader

Collaborators

- L. HENRIQUE, Assistant Professor (IPP, Porto)
- O. INÁCIO, Invited Professor (IPP, Porto)
- M. MOREIRA, Assistant Professor (IPS, Setubal)
- M. PAULINO, Invited Professor (IPL, Lisboa)
- R. SAMPAIO, Assistant Professor (ENIDH, Lisboa)
- R. TABORDA, Assistant Professor (FCL, Lisboa)

Technicians

- A.P. ANASTACIO, laboratory technician

Funding

	$\times 10^3$ PTE
Research Projects:	13 508
Total:	**13 508**

Publications

Book chapters:	1 in press
Journals:	1 and 4 in press
Proceedings:	8
Conf. Communications:	2
Internal Reports:	3

Simulation and identification of complex dynamic systems (ROTDYN)

J. Antunes, M. Moreira[1], H. Pina[2]

Objectives

This project is developed in collaboration with two portuguese engineering schools, IST and IPS, in the framework of a PhD thesis. It has recently been funded under the Sapiens programme. Our aim is to apply stochastic computational methods to dynamical problems of interest to industrial components. As test problems, we work on rotor-flow and axial-flow coupled vibrations, which are problems involving strong non-linear effects.

Results

During 2001 we extended our Galerkin/spectral theoretical formulation of immersed rotors to deal with orbital motions. The theoretical results are currently being confronted with experiments. Furthermore, the theoretical analysis of this non-linear system highlighted the existence of post-stable limit cycles. Most of the know-how acquired during the full extent of this project has now been condensed in a extensive book chapter, edited by WIT Press, currently in print.

Furthermore, two new interesting problems of practical significance, in particular for nuclear facilities, have been theoretically addressed during 2001: The flow-coupled vibrations of stored fuel racks, and the squeeze-film dynamics of vibrating structures subjected to axial flow, such as valves. The results of these developments were recently published.

Experimental set-up for rotor-flow interaction tests.

Published, accepted or in press work

1. M. Moreira, J. Antunes, H. Pina, An improved model for rotors subject to dissipative annular flows, *Journal of Fluids and Structures* (in print).
2. M. Moreira, J. Antunes, H. Pina, A symbolic-numerical method for nonlinear rotor dynamics under fluid confinement, *International Journal of Nonlinear Science and Numerical Simulation* (in print).
3. M. Moreira, J. Antunes, H. Pina, Analysis of nonlinear motions of immersed rotors using a spectral/Galerkin approach, ICSV8-*Eighth International Congress on Sound and Vibration*, Hong-Kong, China, July 02-06, 2001.
4. M. Moreira, J. Antunes, A simplified linearized model for the fluid-coupled vibrations of spent nuclear fuel racks, *ASME PVP - Symposium on Flow-Induced Vibration*, Atlanta, July 22-26, 2001. Accepted for publication in *Journal of Fluids and Structures.*
5. J. Antunes, P. Piteau, A nonlinear model for squeeze-film dynamics under axial flow, *ASME PVP - Symposium on Flow-Induced Vibration*, Atlanta, July 22-26, 2001. Accepted for publication in *Journal of Fluids and Structures.*
6. M. Moreira, H. Pina, J. Antunes, Nonlinear vibrations of asymmetrically supported vertical rotors under annular flow confinement: *Theoretical Results, Fluid Structure Interaction 2001*, Halkidiki, Greece, 26-28 September 2001.
7. J. Antunes, Dynamics of rotor/flow coupled systems, *Book chapter in* Flow Induced Vibrations*, P. Anagnostopoulos (Ed.)*, WIT Press (in print).

[1] Instituto Politécnico de Setubal (IPS)
[2] Instituto Superior Técnico (IST), Lisboa.

Dynamical modelling of nonlinear systems (EXCITE)

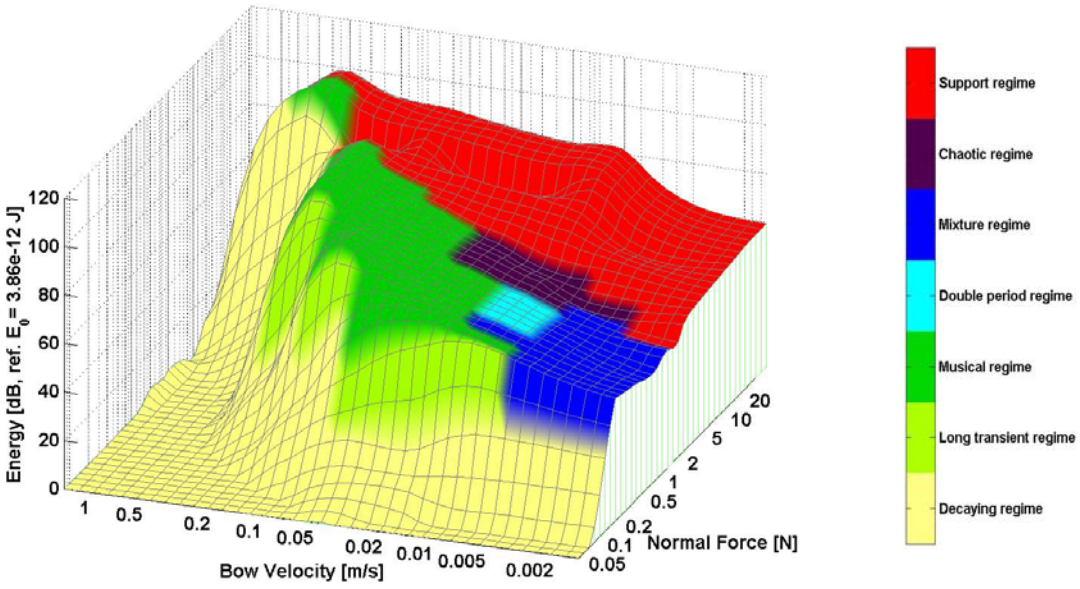
J. Antunes, O. Inácio [1], L. Henrique[1]

Objectives

This project of fundamental nature has recently been funded under the Sapiens programme. A PhD thesis was started in connection with this work. It is an international co-operative effort to develop theoretical methods and numerical techniques to deal with strongly non-linear dynamical problems. Here, computational techniques are developed to study a paradigmatic and difficult problem in music acoustics: the bowed string. The motivation of this work is that such techniques can then be adapted to many related vibrational problems in industrial facilities.

Results

During 2001 we extended our computational method for simulating the dynamics of dispersive wave-propagating systems, such as bowed bars. This enables simulation of the stick/slip behaviour due to frictional forces, as well as self-excited vibrations and bifurcation phenomena. These techniques can be applied effectively to important friction-dominated problems, such as automotive brake devices. Eperimental validation of our simulation results is currently being performed. Furthermore, we started to develop optimisation techniques for dynamical systems, with considerable success.



Dynamical regimes and vibration energy of a bowed bar as a function of the bow normal force and tangential velocity.

Published, accepted or in press work

1. O. Inácio, L. Henrique, J. Antunes, Influence des modes de torsion sur les régimes dynamiques des cordes frottées, *Rencontre sur les instruments à cordes : Entre facture et musique*, Universidade Nova de Lisboa, Portugal, Avril 20-21, 2001.

2. O. Inácio, L. Henrique, J. Antunes, Dynamical analysis of bowed bars, ICSV8-*Eighth International Congress on Sound and Vibration*, Hong-Kong, China, July 02-06, 2001.

3. J. Antunes, L. Henrique, O. Inácio, Aspects of bowed-string dynamics, Invited paper at the *17th Int. Congress on Acoustics (ICA 2001)*, Roma, 3-7 September 2001.

4. L. Henrique, J. Antunes, J. S. Carvalho, Design of musical instruments using global optimization Techniques, *International Symposium of Musical Acoustics (ISMA 2001), Perugia*, 10-14 September 2001.

[1] Instituto Politécnico do Porto (IPP)

Dynamical modelling of geological inclusions (GEOMODELS)

F. Ornelas [1], R. Taborda[1], J. Antunes

Objectives

This project is being developed in co-operation with the Geology Department of FCL and several international partners. It is funded under the Sapiens research program. Our contribution consists on the development of a theoretical model to predict the complex rotations of geological inclusions as a function of the shear motions of the enclosing matrix, which can be modelled as a fluid. This problem is important when trying to understand geological patterns and their history.

Results

During 2001 we pursued the development of a finite element model of the flow/inclusion interaction, which is based on a classic Stokes flow approximation, with moving boundary-conditions. This computational model is currently being refined and validated using the experimental work performed at FCL/LATEX. Results of this work will be published soon. Future work includes the extension of our computational model to more complex inclusion shapes.

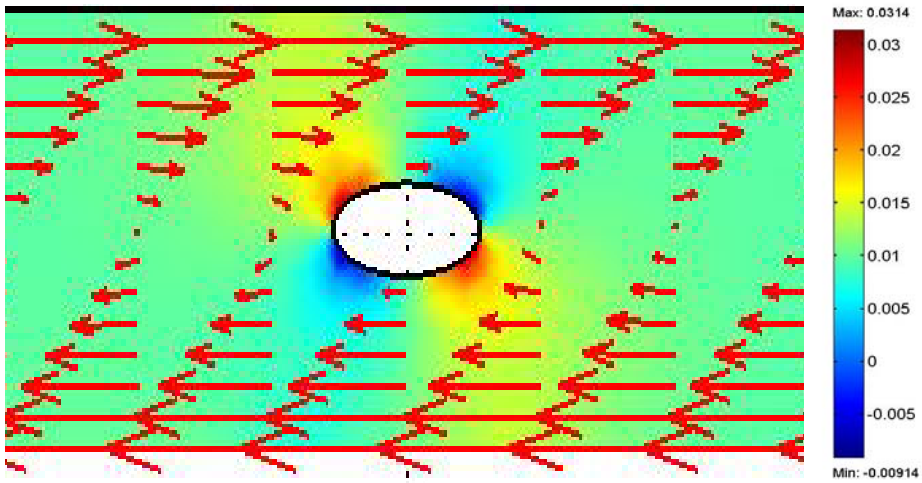


Fig. Velocity and pressure flow fields around an elliptical inclusion under a confined Stokes shear flow.

[1] Faculdade de Ciências de Lisboa (FCL) / Structural Geology (LATEX)

Optimisation of rotor balancing

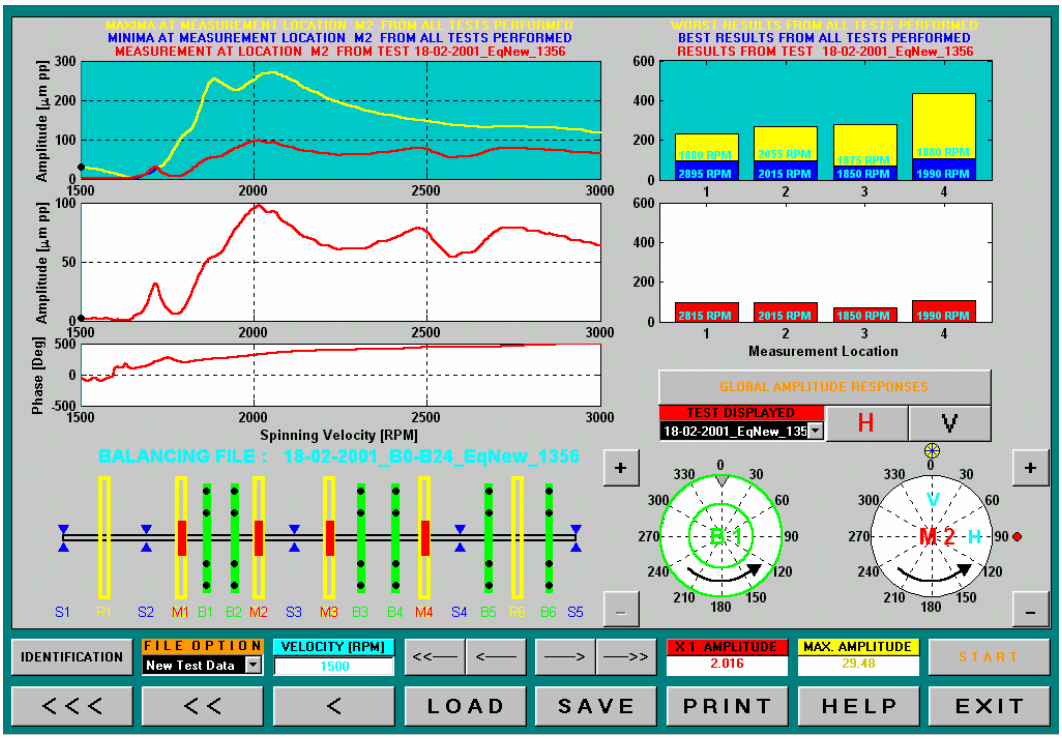
J. Antunes, M. Paulino [1], J. Soares[2]

Objectives

Balancing is one the most common and important tasks currently performed on all rotating machinery. Because the traditional way of rotor balancing (using influence coefficients and many test runs) is very time-consuming and costly, our aim is to optimise these operations. Therefore, we depart from standard procedures and are developing a new method which will enable balancing of flexible rotors at a fraction of the cost and time. If successful, our approach will be of great interest for many industrial facilities. However, this is definitely not an easy task. This project has been porsued under contract for the portuguese power supplier (EDP/CPPE). A PhD thesis is being prepared in connection with this project.

Results

During 2001 an improved test rig (SPIN/2) has been designed and built, in order to test the performance of our innovative approach for balancing. A user-friendly computer program for field balancing, which implements current methods as well as our new approach, is under development. Some preliminary theoretical and experimental results have been published. However, further tests will be needed during 2002, in order to assert the validity range of the new balancing method. Furthermore, we believe that more robust balancing results can be produced by introducing additional constraints in the balancing procedure. Such approach will also be pursued in the near future.



Sample page of the balancing program SPIN, developed by ITN/ADLfor EDP/CPPE: Experimental vibratory levels respectively before (yellow) and after (red/blue) balancing.

Published, accepted or in press work

1. J. Antunes, M. Paulino, Balancing of flexible rotors from a single additional test, *18th Canadian Congress of Applied Mechanics (CANCAM 2001)*, St. John's, Canada, June 3-7, 2001.

2. M. Paulino, J. Antunes, Optimal balancing of flexible rotors: 4 – Preliminary tests of a new balancing method, ITN/ADL Report, May 2001.

[1] Instituto Superior de Engenharia de Lisboa (IPL/ISEL)
[2] Electricidade de Portugal / Companhia Portuguesa de Produção de Electricidade (EDP/CPPE)

Remote identification of impacts

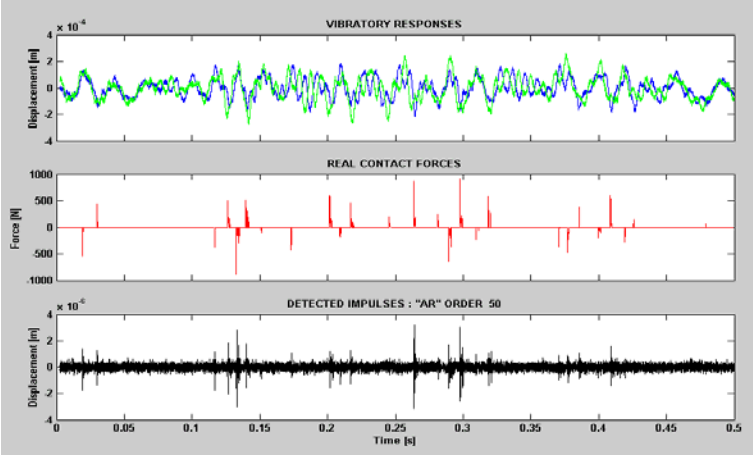
J. Antunes, M. Paulino [1], R. Sampaio[2], L. Borsoi[3], P. Izquierdo[3], P. Piteau[3]

Objectives

This project has been developed for several years, aiming the identification of impact phenomena inside critical components through signal processing of vibratory measurements from transducers at remote locations. This is to avoid placing force transducers under severe environmental conditions (space, temperature, radiation). Such study is of particular significance for condition-monitoring of wear-prone components, such as heat-exchangers, and also as a tool for the analysis of ageing industrial facilities. The later aspect is of particular relevance for nuclear power facilities. This is a very difficult problem, because the interesting signals are often immersed in high noise contamination from many sources.

Results

Our past work in this field mainly concerned about wave-propagation modelling techniques. We also have explored the so-called "blind" identification methods for the remote identification of spiky impact forces. These methods were originally developed in very different fields to enable the identification of impulsive signals without any information on the dynamics of the propagation path. We have extended the original minimum entropy deconvolution algorithm, to enable the effective use of simultaneous response measurements. This technique uses higher-order statistics of the multiple data in order to enable the identification of the impulsive sources. The developed identification method was validated experimentally, with effective results in many practical cases. Although our CEA contract is now completed, we have recently explored several complementary issues, pertaining to the robustness of the identification methods for complex systems. We also recently developed a new detection technique of impact phenomena based on auto-regressive (AR) modelling of the vibratory responses. Many significant publications and an international prize stemmed from this project.



Remote detection of impacts from the noisy vibratory responses of a structure using AR modelling.

Published, accepted or in press work

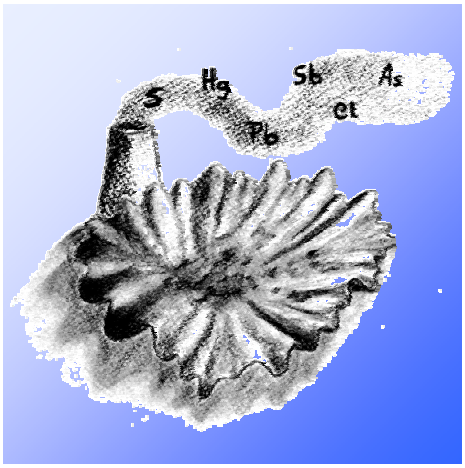
1. J. Antunes, P. Izquierdo, M. Paulino, Blind Identification of Impact Forces from Multiple Measurements, *International Journal of Nonlinear Science and Numerical Simulation*, Vol. 2, pp. 1-20, 2001.

2. J. Antunes, L. Borsoi, P. Izquierdo, P. Piteau, Processus d'identification en vibrations non-linéaires de poutres à chocs, *Société Française de Mécanique*, *Journées sur l'analyse modale expérimentale*, Blois, 29-30 Novembre 2001.

3. J. Antunes, R. Sampaio, Detection d'impacts à distance à l'aide de techniques auto-régressives, ITN/ADL Report, December 2001.

[1] Instituto Superior de Engenharia de Lisboa (IPL/ISEL)
[2] Escola Nautica Infante D. Henrique (ENIDH)
[3] Commissariat à l'Energie Atomique (CEA Saclay/ DM2S/SEMT/DYN)

Atmospheric Elemental Dispersion



Atmospheric Elemental Dispersion

Maria do Carmo Freitas

The aim of this research unit is the evaluation of elemental concentrations in suspension in the atmosphere, the characterization of atmospheric dispersion of chemical elements subjected to local, mesoscale or long-range transport, and its effects or impacts upon the biosphere, including Man. For this, both monitoring (air filtering and deposition sampling) and biomonitoring are used. The field is both a natural application of the potentialities of k_0-INAA and PIXE techniques, which are the historical roots of the unit, and a key-subject in environmental problems. The development of the two nuclear analytical techniques used runs in parallel with their applications. These include the use of the Portuguese Research Reactor (Reactor Sector) and the Van de Graaff accelerator (Physics Sector). The unit activities include four main lines:

A. Monitoring, Biomonitoring, Quality Control, Automation and Data Handling

Polluted areas of Portugal are characterised using lichen transplants, air particulate matter collection, and wet deposition. The data are analysed for factors aiming at identifying emission sources and the spread of elements through the atmopsphere, both locally and by long-range transport. In conjunction to this, data analysis methods use and development are very important due to the multielement nature of the analytical techniques used. This nature makes that specific methods can and are being used and developed to profit the most out of this type of data. To assure the quality of the data used, accuracy and precision studies on both nuclear techniques are being performed both in monitoring and biomonitoring fields, aiming at better understanding differences found in the results for the same element and sample, obtained by the two nuclear techniques. Air particulate matter obtained by different air samplers is compared. The so-called conventional analytical techniques are applied to complement the unit's results. Automation and specific sampling equipment improvement research is carried out. This year (as well as in the previous 2 years), this line is the major source of external financing.

B. Chemical Element Speciation

INAA and PIXE techniques give rise to total contents or concentrations of chemical elements. With this activity we aim at obtaining a better knowledge of the chemical state of the elements to understand better its toxicity towards the environment and the human being. The specific item of chemical element speciaton in atmospheric particles and in biomonitors is a full state of the art field being under development worldwide, almost at request of the Environmental Science Community. presently the unit proceeds with the first steps for the so-called operational and complete (classic) speciation. The first aims at simulating atmospheric conditions for air particulate matter in order to find out the most mobile fraction, the fraction containing the inorganic compounds, and the silicate and organic fraction. The second aims at identifying in lichens the state valence of the chemical element and the compound where it is included.

C. Atmospheric Trace Elements Effects Upon the Biosphere

This subject groups the activity oriented towards the study of the response of biomonitors to general element availability in the environment, which methods and conclusions may also be applied to effects upon Man. Presently the first steps are being given on biomonitoring *versus* epidemiological studies.

D. Training, Services

The unit has a strong component in training. One PhD was completed and two PhDs are ongoing. Two PhDs were initiated this year within activities A and B. Two MScs are being finished to support activity A. Analytical services were also provided to various persons requesting them.

Future Prespectives

In the near future, standard monitoring and integration with biomonitoring will be continued, and stressed by increase of the services component but also through the strengthen of existing colaborations. Data analysis methods, namelly receptor models will be subjected to intensive research and merging to dispersion models is being aimed at. An air sampling prototypes developed is expected to become in use in the next year, and also to become available as an ITN product. Still, line B will become the main research topic, in accordance with the scentific investment policy directives and programs for the next years. Chemical element speciation will receive a special attention by using the low detection limits of k_0-INAA to analyse chemical separation products, but also by investing in new developments in PIXE aiming at in-beam chemical speciation analysis. Relating to line C, the quantification of the impact of trace element upon the biosphere will focus mainly on finding correlations between biomonitor data and health data, as well as stress effects on biomonitors.

Atmospheric Elemental Dispersion - DEA

Research Team

Researchers

- M.C. FREITAS, Principal Researcher, Group Leader
- M.A. REIS, Auxiliary Researcher

Students

- A.P. MARQUES, PhD Student, ITN grant
- S.M. ALMEIDA, PhD Student, FCT grant
- C.H. GOMES, PhD Student, FCT grant
- M.M. FARINHA, PhD Student, ITN grant
- C.J. COSTA, MSc Student, invited
- C. MUSTRA, MSc Student, ITN grant
- O.R. OLIVEIRA, Graduate Student, ITN grant
- M. VENTURA, Graduate Student, ITN grant
- A. VARAJÃO, Undergraduate student, IST
- C. PASSARINHO, Undergraduate student, IST
- I. SOARES, Undergraduate student, IST
- D. CARVALHO, Graduate Student, ITN grant

Technicians

- I. DIONÍSIO, Laboratory Technician
- R. PINHEIRO, Laboratory Technician

Collaborators

- A.M.G. PACHECO, Auxiliary Professor, IST, Technical University of Lisbon, Lisbon
- H.TH. WOLTERBEEK, Senior Researcher, IRI, TUDelft, The Netherlands
- J. de GOEIJ, Senior Professor, IRI, TUDelft, The Netherlands

Funding

	$\times 10^3$ PTE
Research Projects:	18 033
Total:	**18 033**

Publications

Book Chapter:	1 in press
Journals:	4 and 4 in press
Proceedings:	2 and 3 in press
Conf. Communications:	7
Thesis:	1 PhD
	1 Graduation

Biomonitoring, Alternative Biomonitors, Monitoring, Calibration

M.C. Freitas

Objectives

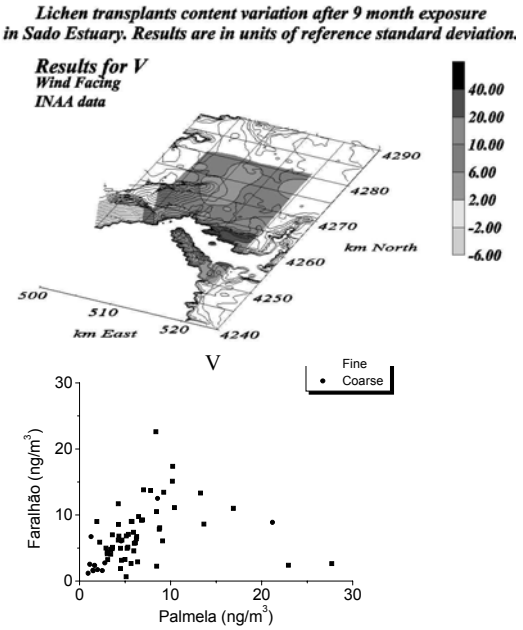
In this activity we aim at interconnecting the works and results performed within the unit, attempting to extract the information, which might be useful for more than one activity, and to correlate the different activities.

Results

Monitoring vs. biomonitoring

Monitoring and biomonitoring was put together in a publication[1] where results on lichen transplants of Sado estuary were studied together with the results on air particulate matter in the same region. Vanadium is an important tracer of the fuel combustion. Fig. 1 shows, for vanadium, the aerosol results in two locations nearby the Setúbal fuel power station, and the results obtained for the lichen transplants in a 15km x 25km area around it. Fig. 1 shows that the concentrations found in both

Fig. 1 Results on lichen transplants and aerosols in Sado estuary.



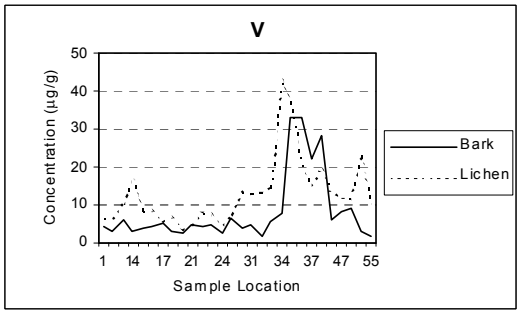
stations are correlated; therefore the major source of this element is the same: the fuel power station. Faralhão reveals higher concentrations as expected, since it is located closer the fuel power station. The lichen results reinforce these conclusions, pointing out clearly the source. Concerning the Sado estuary, factor analysis was done[2] aiming at identifying emission sources. Sea spray, a soil component and industrial emissions of complex origin were identified.

In situ lichens vs. transplanted

An overview was made[3] where a few examples of air monitoring with transplanted and native organisms were presented to illustrate their usefulness. Lichen transplants were found to respond quickly and effectively to the elemental emission sources, which they reflect, though their position relative to the prevailing winds may turn into differences in source indication. The response of indigenous lichens appeared somewhat less clear.

Alternative biomonitors

Fig. 2 Olive tree bark *vs. in situ* lichens.



Studies with olive-tree bark[4] have shown that it could be an interesting alternative to common epiphytes, at least in southern Europe (see Fig. 2).

Monitoring

The directive 96/62/CE concerns the evaluation of the air quality in European Union. It foresees legislation for cadmium, arsenic, nickel and mercury to complement the already legislated lead. The group has large experience in determining these chemical elements as well as lead, using the nuclear techniques INAA and PIXE to analyse the airborne particulate matter. It is reported[5] the possibilities of these two techniques to analyse the chemical elements subjected or to be subjected to regulation. Data on these elements are reported for a few places in Portugal, and compared to existing regulations.

Calibration

k_0 Standardized neutron activation analysis is very sensitive to reactor parameters. Therefore each time the reactor configuration changes these should be re-determined. Table I shows the improvement in the analysis of lichen reference material IAEA 336 obtained by introducing the reactor parameters determined for the 2001 configuration modification. These new data were introduced in the analyses performed to up-date the results.

Table I Modification of results by introducing *f* and α determined after last RPI configuration change

Element	Cert. value	Before	After
Br	12.9	11.2	12.3
Fe	430	467	425
K	1840	1994	1817
Zn	30.4	31.3	28.8
Cr	1.06	1.4	1.2

Published, accepted or in press work

1. C.J. Costa, A.P. Marques, M.C. Freitas, M.A. Reis, O.R. Oliveira, A Comparative Study for Results obtained using Biomonitors and PM10 Collectors in Sado Estuary, *Env.Poll.*, in press.
2. M.C. Freitas, M.A. Reis, A.P. Marques, H.Th. Wolterbeek, Use of Lichens Transplants in Atmospheric Deposition Studies, *J. Radioanal. Nucl. Chem.* 249 (2) (2001) 307-315.
3. M.C. Freitas, A.M.G. Pacheco, A.P. Marques, L.I.C. Barros, M.A. Reis, *Application of Nuclear Analytical Techniques to Environmental Studies, Proceedings of the 16th International Conference Application of Accelerators in Research and Industry'2000, eds. Jerome L. Duggan and I. Lon Morgan, American Institute of Physics Publ.*, College Park MD, USA, 2001, pp. 508-511.
4. A.M.G. Pacheco, M.C. Freitas, M.A. Reis, M.G. Ventura, L.I.C. Barros, An Assessment of the Response of Tree Bark and Epiphytic Lichens to Elemental Availability at Ground Level through Nuclear Techniques, *47th Annual Radiochemical Measurements Conference*, Honolulu, HI, November 4-8, 2001.
5. M.C. Freitas, M.M. Farinha, S.M. Almeida, C.J. Costa, M.A. Reis, O.R. Oliveira, A.M.G. Pacheco, M.G. Ventura, Measurement of Lead, Cadmium, Arsenic, Nickel and Mercury in PM10 and PM2.5 by means of INAA and PIXE, *3rd International Symposium on Nuclear Analytical Chemistry (NAC-III)*, Halifax-NS, Canada, June 11-14, 2001. Submitted to *J. Radioanal. Nucl. Chem.*

Wind Effects and Quality Control of Biomonitoring Data

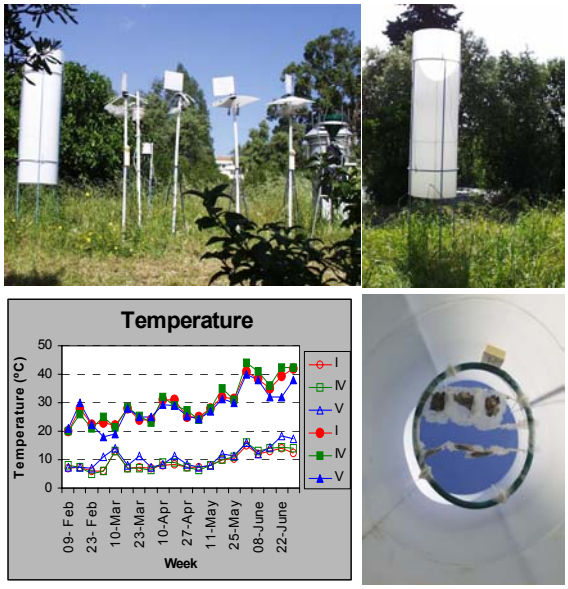
A.P. Marques, M.C. Freitas, M.A. Reis, O.R. Reis, H. Th. Wolterbeek[1], J. de Goeij[1]

Objectives

This is basically the PhD work of A.P. Marques and is divided in three parts: comparison of results given by INAA and PIXE and the effect of the sample particle grain size on their outcomes; analysis of the enhancement by wind direction differential exposure of biomonitors; and the transplant case study in Sado estuary.

Results

Fig.1 Study at ITN campus, detail of the set-up and temperature measurements.



In the case of Sado estuary the Monte Carlo aided Target Transformation Factor Analysis (MCTTFA) method from IRI-TUDelft has been applied to the data and the outcomes are presently under evaluation.

For further study of wind effects, during 2001, a complex set-up experiment for studying lichen transplants was carried out at ITN campus. Transplants were placed in such way that three conditions were established: *i*) free elemental influx, *ii*) horizontal influx only, thereby discriminating wind-directional positioning of the transplant, and *iii*) vertical influx only (Fig. 1).

Several studies (light, turbulence and temperature measurements) were performed to build the vertical influx only lichen transplant support.

For INAA and PIXE comparisons a detailed study started with IAEA-336 lichen reference material consisting basically of two steps: separation of IAEA-336 (IAEA-336 has a grain size below 125 μm) in different fractions. Sieves of polyethylene material were built with 3 different nylon nets of 64 μm, 41μm, and 20μm. Grinding was made whenever necessary. The fractions were analysed by PIXE and INAA, the results were compared and they are shown in Fig. 2.

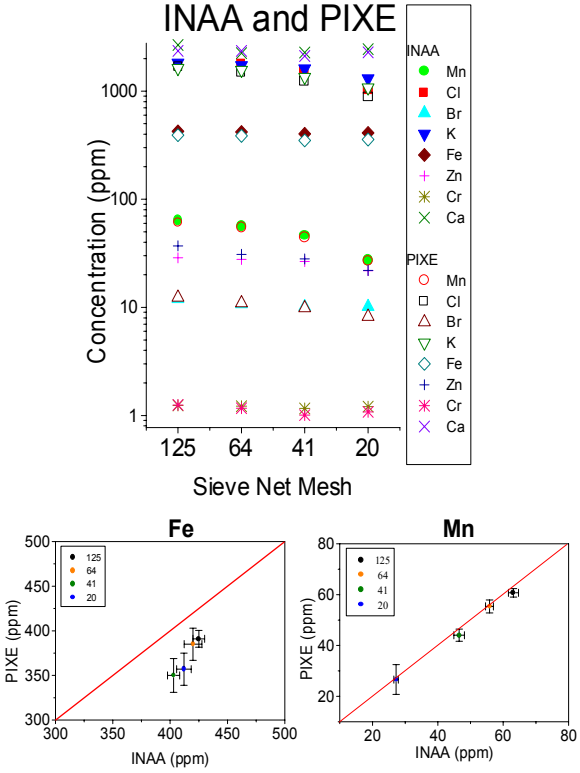


Fig. 2 Results obtained by PIXE and INAA. Comparison of different fractions of lichen material.

Published, accepted or in press work

1. M.C. Freitas, M.A. Reis, A.P. Marques, H.Th. Wolterbeek, Use of Lichens Transplants in Atmospheric Deposition Studies, *J. Radioanal. Nucl. Chem., Articles*, 249 (2) (2001) 307-315.

[1] Radiochemistry Department, IRI-TU Delft, The Netherlands.

Use of Tree Bark as an Air Pollution Biomonitor

M.C. Freitas, A.M.G. Pacheco[1], M.A. Reis, L.I.C. Barros[1], M.G. Ventura, O.R. Oliveira

Objectives

The indication that tree bark could be analysed in the same way as lower epiphytes led to an increasing interest in that material as an air pollution monitor. Tree bark has been used far less than lichens, bryophytes or non-lichenised fungi: bark studies are truly scarce and mostly related to environmental acidification. Nevertheless, biomonitoring with bark could have several advantages that should not be overlooked, meaning: an availability of biological material year-round, an easier identification and sampling when compared with lichens or bryophytes, and an ubiquity of some genera that makes it feasible to survey extensive areas without endangering any species or putting them in short supply. In our group bark of olive tree (*Olea europaea*) – a common, widespread species in Mediterranean and Southern-Europe countries – is being studied in order to establish its availability to biomonitoring heavy metals present at atmosphere.

Results

A grid was established covering the whole Portuguese territory. Due to the high industrial asymmetry between the coast and the interior of the country two different grids were defined: along the Atlantic coast (a 10 x 10 km grid) and in the interior (a 50 x 50 km grid). A total of 228 sampling squares were thus defined. The olive bark samples were collected together with the lichens ones within each square and the nearest possible of the centre. Samples were analysed by INAA and PIXE. Prior to analysis, the bark material was cleared from lichens and dust, rinsed under deionised water for 30 s, freeze-dried, ground in a teflon™ mill.

Figure 1 represents the study area, which comprehends the north and centre regions in littoral of Portugal.

Results for As and V determined by INAA and Pb and Ni determined by PIXE are presented in Figure 2. litoral-north and litoral-centre, showing the 10x10 km grid for biomonitoring.

Values in bark ranged between 0.0645 μg/g and 4.94 μg/g for As; 0.444 μg/g and 35.7 μg/g for V; 0.976 μg/g and 14.6 μg/g for Ni and 3.28 μg/g and 72.2 μg/g for Pb.

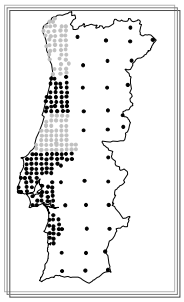


Fig.1 Map of the two study areas (grey) in Portugal:

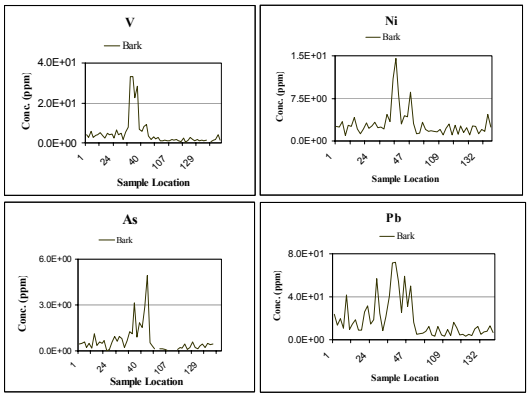


Fig.2 As, V, Pb and Ni levels in *Olea europea* bark at North and Litoral centre of Portugal.

Published, accepted or in press work

1. A.M.G. Pacheco, M.C. Freitas, L.I.C. Barros, R. Figueira, Investigating Tree Bark as an Air - Pollution Biomonitor by Means of Neutron Activation Analysis, *Journal of Radioanalytical and Nuclear Chemistry* 249 (2) (2001) 327-331.

2. A.M.G. Pacheco, L.I.C. Barros, M.C. Freitas, M.A. Reis, C. Hipólito, O.R. Oliveira, An Evaluation of Olive Tree Bark for The Biological Monitoring of Airborne Trace-Elements at Ground Level, *Environmental Pollution*, in press.

3. M.C. Freitas, M.A. Reis, A.M.G. Pacheco, L.I.C. Barros, Biological Monitoring of Environmental Contaminants - Established Systems and Potential Newcomers, *Eurasap Workshop on Air Pollution & the Natural Environment: Biological Monitoring*, Sofia, Bulgaria, April 25-27, 2001.

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Atmospheric Monitoring at the S. João da Talha Urban Waste Incinerator

S.M. Almeida, M.M. Farinha, A.P. Marques, O.R. Oliveira, I. Dionísio, R. Pinheiro, M.A. Reis, M.C. Freitas

Objectives

Three years ago an incinerator started to burn domestic urban waste (S. João da Talha Urban Waste Incinerator) of the metropolitan area of Lisbon (Amadora, Lisbon, Loures and Vila Franca de Xira). This work is associated with the monitoring studies of this structure. This activity aims at monitoring the air quality (in a discontinuous way) at the surroundings of the incinerator, as well as its effects on the neighboring vegetation. The first is done under contrac with IDAD (Institute for Environmental Development, Aveiro University) aiming at the determination of a few metals in air particulate matter; and the second under direct contract with ValorSul (Incinerator's owner) aiming at determining a few trace elements.

Results

IDAD's contract foresees the monthly report in PM2.5 and PM10 of the following metals: lead (twice a week), chrome, copper, manganese, nickel, arsenic, cadmium and mercury (once a week), and potassium, antimony, vanadium and zinc (one time every two weeks). The sampling refers to: three stations for air particulate matter (Bobadela, S. João da Talha and Póvoa de Santa Iria) using Gent samplers, and two stations for vegetation (Santa Iria de Azóia and close to the Incinerator).

This contract is now finishing its 3rd year, allowing comparison of PM10 mass concentration and PM10 lead concentration with the Portuguese (EU) legislation. The average of PM10 mass concentration (see Fig. 1)

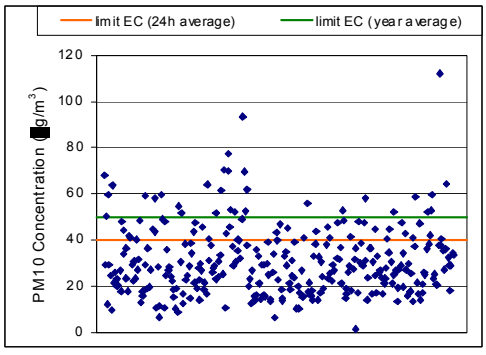


Fig.1 PM10 mass concentration for a 3-year period. Location Bobadela.

for the 3 years is 31 µg/m^3 (lower than the legislated 40 µg/m^3); for 24 h the 50 µg/m^3 was exceeded 28 times (less than the legislated value of 35 times). Concerning lead, under EU and Portuguese legislation, the values are quite low (10 times lower) as compared to the legislated value of 0.5 µg/m^3.

Concerning vegetation, an increase in enrichment factors of mercury, antimony, selenium and euopium was observed during spring and summer in the vascular plant *Conyza*. Fig. 2 shows the evolution of Sb and Hg in this plant along the last 3 years. This might have been the effect of a very local emission source since it was not observed in air particulate matter collected by the time in Póvoa de Santa Iria, close to Santa Iria da Azóia where the enrichment was observed.

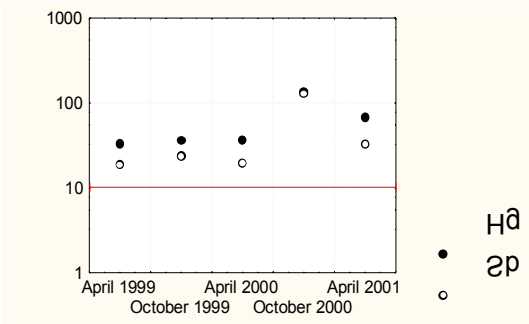


Fig. 2 Increase of Hg and Sb enrichment factor in vascular plant *Conyza*. Location: Sta Iria de Azóia.

Published, accepted or in press work

1. M.M. Farinha, M.C. Freitas, S.M. Almeida, M.A. Reis, *Rad. Phys. Chem*. 61 (3-6) (2001) 659-661
2. S.M. Almeida, M.M. Farinha, M.C. Freitas, C.J. Costa, M.A. Reis, O.R. Oliveira, *Proceedings of the 6th International Conference on Technologies and Combustion for a Clean Environment*, Porto, Portugal, July 9-12, 2001, pp 293-297.
3. M.C. Freitas, M.M. Farinha, S.M. Almeida, S.M., C.J. Costa, M.A. Reis, O.R. Oliveira, A.M.G. Pacheco, M.G. Ventura, *3rd International Symposium on Nuclear Analytical Chemistry (NAC-III)*, Halifax-NS, Canada, June 11-14, 2001. Submitted to Journal of Radioanalytical and Nuclear Chemistry.
4. S.M. Almeida, M.M. Farinha, M.C. Freitas, M.A. Reis, O.R. Oliveira, *47th Annual Radiochemical Measurements Conference*, Honolulu, HI, November 4-8, 2001.

Particulate pollution in an industrial and urban area – ITN Campus

S.M. Almeida, C.A. Pio[1], M.C. Freitas, M.A. Reis

Objectives

The objective of this work is the determination of the elemental and inorganic aerosol composition and the determination of the sources, which contribute to this aerosol. In order to achieve this, knowledge on data quality and representativity must also be obtained.

Results

In January 2001, a new air particulate matter sampling program started, using two Gent collectors, one High Volume sampler (Hi-Vol) and a Berner Impactor. These samplers were placed in a 25m^2 area situated in the ITN campus.

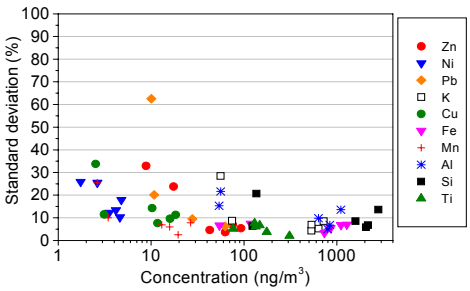
The Gent collectors and the Hi-Vol sampler work during a 24 hours period. Sampling is made twice a week: on Sunday and on a midweek day. These three samplers are connected to the main power by a common timer thus assuring simultaneous work. The impactor works during 72 hours once per season.

Filter loads are measured by gravimetry using a balance with 0.1 µg sensitivity. Elemental analyses are carried out using INAA and PIXE techniques for filters provided by the Gent collectors and Berner Impactor.

Filters provided by the Hi-Vol sampler will be analyzed by chromatographic techniques, for anion determination and atomic absorption for cation determination in Aveiro University.

The data quality and representativity question was addressed by making a precision study, which results relating the element concentrations in filters and the precision obtained are presented in Fig.1.

The mass concentrations obtained by the Gent collectors and the Hi-Vol sampler was also compared to determine the reprodutibility of the two systems (Fig. 2), and finally, the concentrations of the elements determined simultaneously by INAA and PIXE, were also compared to check the accuracy



inbetween the techniques (Fig. 3).

Fig. 1 Precision study.

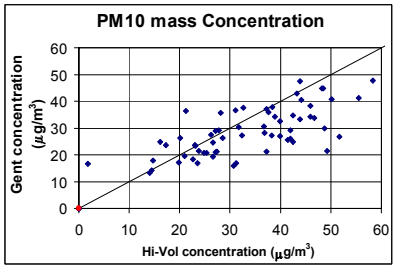


Fig. 2 Comparison between mass concentrations obtained by Hi-Vol sampler and Gent collectors.

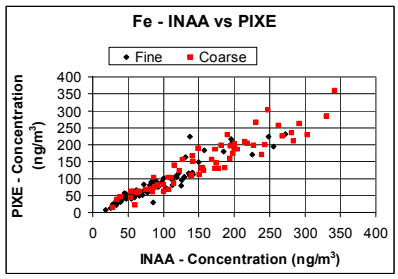


Fig. 3 Comparison between Fe concentrations obtained by INAA and PIXE.

The remaining part of the work carried out this year within this title, makes use of the Berner impactor. While, the Hi-Vol and the Gent samplers collect particles in two different size fractions (a coarse fraction with diameter between 2.5 and 10 µm and a fine fraction with a diameter lower than 2.5µm) The Berner Impactor allows a characterization of eight particles size distributions. In Fig. 4 it is shown the mass distribution in the spring sampling.

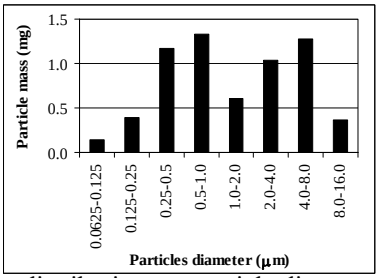


Fig. 4 Mass distribution per particle diameter obtained in the spring sampling.

Published, accepted or in press work

1. S.M. Almeida, M.C. Freitas, M.A. Reis, Quality control of air particulate matter PM10 and PM2.5 by k_0-INAA, k_0 Users Workshop, 23rd-28th September 2001, submitted to *J. Radioanal. Nucl. Chem.*.

[1]Departamento de Ambiente e Ordenamento, Universidade de Aveiro, 3810-193, Aveiro, Portugal

Data Analysis, Source Identification and Long-range Transport

M.A. Reis, O.R. Oliveira, C. Costa, M.C. Freitas, L.C. Alves[1,2], E.M.C. Rita[1,2], F. Rodrigues[3], P. Fialho[3], C.A. Pio[4], J.C. Soares[1,2]

Objectives

Within the sub-subject of data handling there are two important points being addressed: the interpretation of multielement data in term of emission sources, and the application of multielement data to the study of long-range transport processes.

Results

In the framework of long-range transport, data from Faralhão and Azores Islands were compared in a preliminary approach. Results showed that correlations could be found for sulphur data and also for other elements. These results do point the need for further exploitation of this approach.

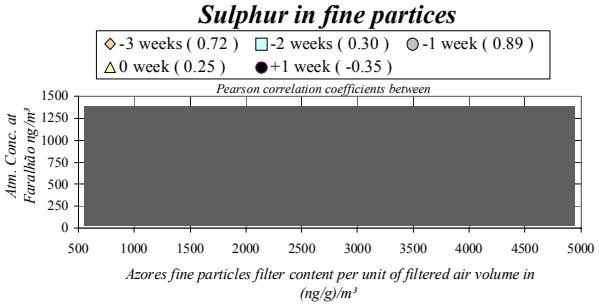


Fig. 1 Faralhão (Nuclepore filters) *versus* Azores (thick quartz filters) data for sulphur in fine particles. Sulphur concentration in the quartz filters normalized to the sampling volume is nearly proportional to the expectable sulphur concentration in Azores atmosphere. The correlation identified for a one-week delay is interpreted as an indication of transport of sulphur from Portugal to Azores, one week being taken for that.

Concerning the interpretation of multielement data the problem to be solved is the interpretation of multielement data in terms of emission sources. Presently used standard airborne particle monitoring methods require lengthy campaigns to obtain large datasets necessary to infer about the main sources affecting a given receptor location. Furthermore, some methods used (eg. factor analysis) have problems to cope with highly variable data, as is the case in airborne particle data samples.

In the work that is presently in progress a new method was established which allows obtaining information on possible sources influencing the atmosphere out of, if necessary, a single airborne particle sample, by making use of multielement data.

This method is named Element Constant Ratio Analysis method (ECRA) and up to now only the theory and a first application was made and presented in 1. Besides this, an extensive paper presenting the theory and applying it to data obtained in one year sampling at two experimental stations is being finished for publication.

In this paper the method is applied to data obtained by using the nuclear analytical techniques PIXE and INAA to analyse PM2.5 and PM10-PM2.5 particles collected onto Nuclepore filters using Stacked Filter Units. Sampling was carried out between December 1997 and September 1998, in two stations located in the industrial region of Setúbal, 30 km to the South-East of Lisbon. Data obtained for week averages of airborne concentration of 29 elements: Na, Al Si, S, Cl, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Br, Mo, Sb, Cs, La, Ce, Sm, Hg, Pb and U, is presented. Source apportionement is restricted to the subset consisting of: V, Cr, Mn, Co, Ni, Cu, Zn, As, Se, Mo, Sb, Hg and Pb, due to the fact that in its stage, the method requires the *a priori* knowledge of the profiles of the emission sources.

Published, accepted or in press work

1. M.A. Reis, Biomonitoring and Assessment of Atmospheric Trace Elements in Portugal – Methods Response Modelling and Nuclear Analytical Techniques, *Delft University of Technology PhD Thesis, Delft University Press*, Delft, The Netherlands, 2001.

2. M.A. Reis, O.R. Oliveira, L.C. Alves, E.M.C. Rita, F. Rodrigues, P. Fialho, C.A. Pio, M.C. Freitas, J.C. Soares, Comparison of Continental Portugal and Azores Islands Aerosol during a Sahara Dust Storm, *Nucl. Inst. and Meth. in Phys. Res* , In press.

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[2] CFNUL, Av Gama Pinto nº 2, 1699 Lisboa, Portugal
[3] Department of Agrarian Sciences of the University of the Azores, Azores, Portugal
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Simple Unit for Collecting Aerosols - URSA

O.R. Oliveira, S.M. Almeida, C. Hipólito, R. Pinheiro, M.A. Reis, M.C. Freitas

Objectives

Within DEA, direct monitoring has been made using Gent samplers provided by IAEA and used for aerosol sampling. The practice in using this kind of samplers revealed some small practical operational problems. Besides this, their internacional manufacturers have shown to have problems in providing equipments (even though the market is not big, or may even be by this same reason). Based on its experience in prototipes for automatic sampling (see previous reports), the unit started to build its own sampling unit and at the same time improved some details.

Results

The ITN Simple Unit for Collecting Aerosol– short name URSA – the two principal blocks shown in Fig. 1 and Fig. 2, is a system designed to collect aerosol particles and some gases, which has some degree of energetic autonomy and is planned to avoid dew point conditions in the filtering unit. These three last characteristics do not exist in the Gent sampler.

In the URSA the pumping subsystem was highly improved in order to achieve a higher electric efficiency. The mechanical structure and the methods of component assembling are also diferent and allow a final sampling unit with dimensions of 40x30x25 cm (Length x Depth x Height). The electric subsystem is DC based, therefore enabling autonomous operation if a battery is used. The autonomy time depends on the battery. The system is designed to allow coupling a normal lead battery, whose charging is then assured if the system is connected to the power line. A normal car battery will provide autonomous operation for time periods up to 5 hours.

Fig. 1 View of the URSA main part.

Fig.2 View of the URSA and PM10 separator.

A new PM10 separator (shown in Fig. 2) was also designed to allow functioning at lower fluxes, compatible with a true PM2.5 separation of the NILU Stacked Filter Units, when used with Nuclepore 8µm pore filters. The design of the Gent PM10 separator aims at a PM2.2 separation in the SFU system.

The extraction of the PM10 top (right piece in Fig. 2) is easy, which simplifies the access to the PM10 separation plate that should be freshly greased before every use.

The mounting in the field and the preparation procedures of the PM10 separator are also simplified.

In Fig.3 the first results of the comparison of the URSA separator and the Gent separators are presented.

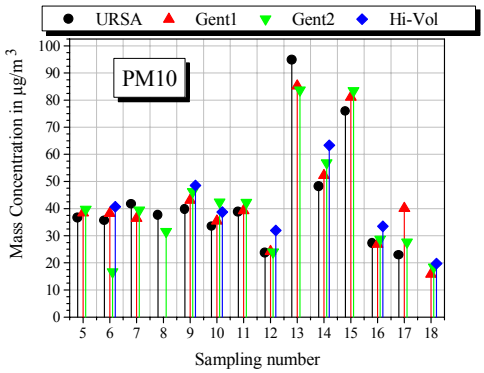


Fig. 3 Comparison of PM10 concentrations obtained by using the new URSA PM10 separator coupled to a Gent pumping system, two Gent systems and one High volume sampler. Along the experiment, small changes were introduced in the URSA separator in order to eliminate some systematic deviations determined in the first measurements.

Trace Metal Operational Speciation in Reference Materials

C. Hipólito, M.C. Freitas, M.T. Vasconcelos[1]

Objectives

The main objective is testing an operational speciation procedure using an extraction certified sediment reference material for future application to aerosol samples. Along the tests ensurance of good reproducibility and accuracy was an objective always present.

Results

Operational speciation is one of the most popular schemes of sequential extraction as was verified during the conference in GSF in Munich[1] related to the subject, where the PhD student was present. The sample is treated with a succession of reagents, of increasing harshness, designed to release metals associated with different phases. Because a large number of different sequential extraction schemes have been used, the Commission of the European Communities, Community Bureau of Reference (BCR) recently devised a simple, three stage sequential extraction protocol for operational speciation of trace metals in attempt to harmonise methodology throughout the EU, and to improve comparability between laboratories.

Table I - BCR sequential extraction procedure

	Reagent	Nominal target phase(s)
1	0.11 mol l^{-1} CH_3CO_2H	Exchangeable, water and acid-soluble
2	0.10 mol l^{-1} NH_2OH, adjusted to pH 2 with HNO_3	Reducible (e.g. iron /manganese oxides)
3	8.8 mol l^{-1} H_2O_2, followed by 1.0 M $CH_3CO_2NH_4$	Oxidizable (e.g. organic matter and sulfides)

We applied the BCR sequential extraction procedure shown in Table I to the reference material CRM 601 (material certified for the EDTA – extractable contents of Cd, Cr, Ni, Pb and Zn in sediment following a three-step sequential extraction procedure). The content concentrations in the extractable solutions were determined by atomic absorption spectroscopy in the case of the elements Cd, Ni, Pb and Zn and by graphite furnace atomic absorption spectroscopy for Cr and compared with the certified values. Results are shown in Figs. 1-3.

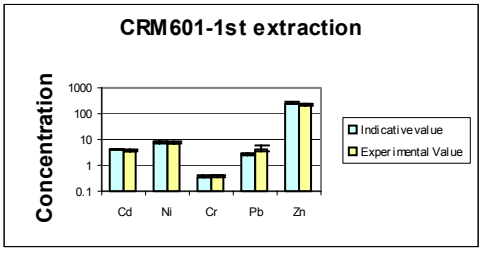


Fig. 1 – Concentration (ppm) of Cd, Ni, Cr, Pb and Zn in first extraction step; results obtained by EAA

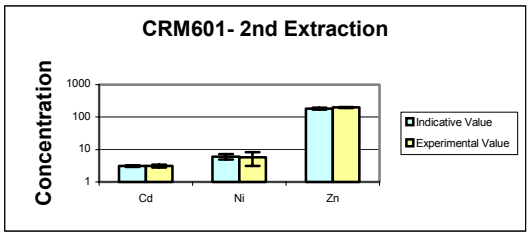


Fig. 2 – Concentration (ppm) of Cd, Ni and Pb in the second extraction step; results obtained by EAA.

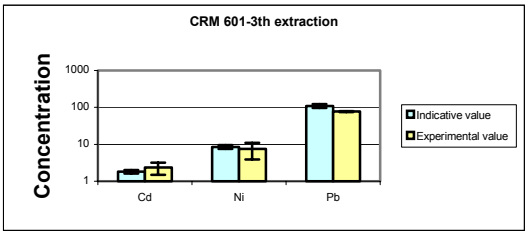


Fig. 3 – Concentration (ppm) of Cd, Ni and Ob in third extraction step; results obtained by EAA.

Quality control involves comparison of results obtained by EAA and INAA. Because only solid samples can be analysed by neutron activation and solutions from extraction procedure are in liquid form we thought in possible ways to pre-concentrate solutions. Some sensibility tests were performed. A few millilitres of a 1000-ppm reference solution of the element to be analysed was evaporated and irradiated. The value obtained by k_0-method was compared to the expectable value.

Analysis of reference material NIST 1633a by INAA and EAA is one of the objectives of the quality control strategy. This is coal Fly Ash sediment that has a matrix similar to that of the real samples and the same values of concentration levels of the analyte certified. We analysed coal fly ash samples by neutron activation analysis (see Fig. 4).

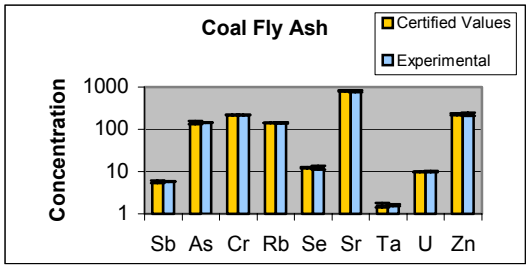


Fig. 4 – Concentration (ppm) of trace elements in coal fly ash. Comparison with the certified values.

1 LAQUIPAI, Departamento de Química, Universidade do Porto

Arsenic Speciation of Lichens

M.M. Farinha, Johannes T. Van Elteren[1], Zdenka Slejkovec[1], M.C. Freitas

Objectives

Metals are present in the atmosphere but only a few of their compounds are toxic, so it is important to know the metal species present. The main aim of this work consists in arsenic speciation of lichen samples from the national survey held in 1993 in order to identify and quantify the arsenic chemical forms. The selection of lichen samples for speciation is based on the information given by the Monte Carlo Aided Target Transform Factor Analysis (MCTTFA) published in 1997.

Results

Fig.1 shows the total concentration in arsenic obtained in the national survey of 1993.

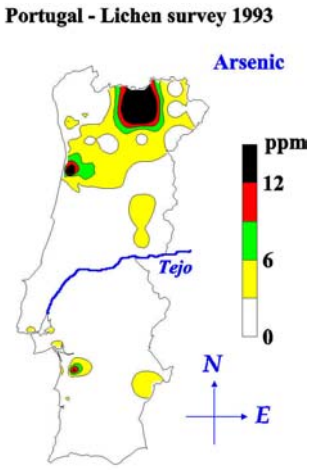


Fig. 1 Total concentration of arsenic

The selection of samples to be analysed by HPLC-HG-AFS was done comparing the results obtained by MCTTFA and the total concentration of arsenic. Thirty samples were chosen: 10 with the highest arsenic concentrations, 10 with lowest and the other 10 in-between. Speciation analysis was done in the Jozef Stefan Institute in Ljubljana (Slovenia).
Lichen speciation consists in two steps: extraction and chromatographic analysis.

The extraction procedure consists in the following steps:

- Solution with MQ water
- Freezing in nitrogen
- Shaking
- Centrifugation
- Filtration
- Drying

The schematic diagram in Fig. 2 can represent all process of analysis.

The compounds on Table I are being determined.

Table I Arsenic compounds obtainedin lichens by chromatographic analysis.

Anionic compound	**Abbreviation**	**Formula**
Arsenous acid	As(III)	H_3AsO_3
Arsenic acid	As(V)	H_3AsO_4
Monomethylarsonic acid	MMAA	$CH_3AsO(OH)_2$
Dimethyarsine oxide	DMAA	$(CH_3)_2AsO(OH)$

Cationic compound	**Abbreviation**	**Formula**
Arsenobetaine	AsB	$(CH_3)AS^+CH_2COOH$
Arsenocholine	AsC	$(CH_3)AS^+CH_2CH_2OH$
Tretamethylarsonium ion	TETRA	$(CH_3)_4As^+$
Trimethylarsine oxide	TMAO	$(CH_3)_3As^+OH$

Results for cromatography are been made in Jozef Stefan Institut.

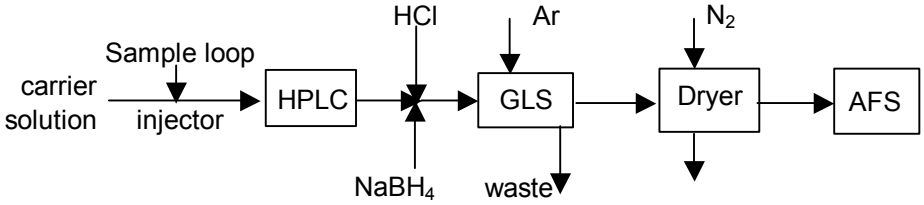


Fig. 2 Schematic diagram of HPLC-HG-AFS.

[1] Jozef Stefan Institut, Ljubljana, Slovenia.

PM10 generalized Equivalent Constant Availability in an Industrialized Area

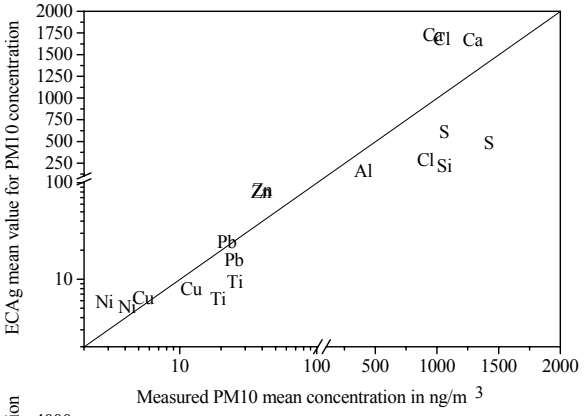
M.A. Reis, A.P. Marques, C. Costa, O.R. Oliveira, R. Pinheiro, I. Dionísio, M.C. Freitas

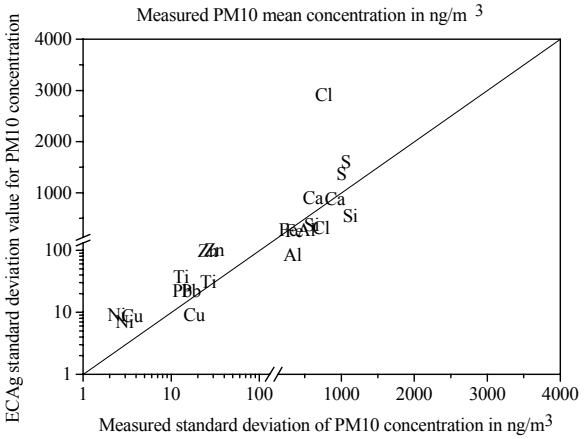
Objectives

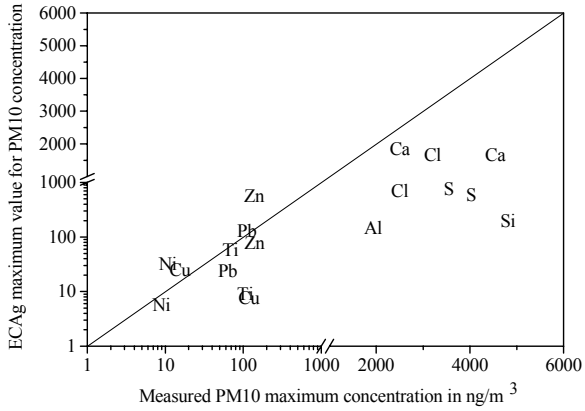
In order to generalize the use of biomonitoring, it is necessary to connect biomonitoring data to other well known environmental parameters. In [1] it was proposed that this could be obtained by accessing the annual variability of biomonitors content. This approach provides simultaneously a way to understand the effects of atmospheric trace elements upon living organisms and a way to infere levels of the atmospheric concentrations out of biomonitoring data. This sub-subject of main line C aims at proving that the proposed approach can be applied to data other than that used for obtaining the calibration parameters. In paralel it is expected that the methods developed may also be used to establish new methods for risk assessment of the human populations or ecosystems stress conditions.

Results

The calibration coefficient for PM10-ECAg were applied to the data from lichens *Parmelia sulcata* transplants exposed in two stations located in the industrial region of Setúbal, 30 km to the South-East of Lisbon, and for which there is also PM10 data. As is presented in [1], the PM10-ECAg parameters are biological biased parameters for the PM10 concetrations statistical distributions. In the figures presented here, the PM10-ECAg mean, maxima and standard deviation for Al, Si, S, Cl, Ca, Ti, Fe, Ni, Cu, Zn and Pb are compared to the direct measurement results obtained for the period of exposure of the transplants. The results obtained so far show that PM10-ECAg does provide a good representation of the statistical description of PM10 composition.







1. M.A. Reis, Biomonitoring and Assessment of Atmospheric Trace Elements in Portugal – Methods Response Modelling and Nuclear Analytical Techniques, *Delft University of Technology PhD Thesis, Delft University Press*, Delft, The Netherlands, 2001.

k_0 Epithermal Neutron Activation Analysis

Carlos Costa, M. Carmo Freitas

Objectives

One of the major interests to perform neutron INAA is the ability to determine a large number of elements (some of them at very low concentrations) simultaneously. In Na-enriched samples sodium may increase dramatically the spectra background due to its (n,γ) high cross section, hiding some important small peaks. In order to optimise the Na-induced background increase, Epithermal Neutron Activation Analysis (ENAA) technique is being attempted on biological samples and air particulate matter.

Results

ENAA analysis consists in irradiating the samples under a Cd cover. This element captures almost all thermal neutrons allowing the transmission of neutrons with energies above 0.55 eV, using pre-defined Cd cover cylindrical dimensions. In this way, the activity induced in the sample will be mainly due to epithermal neutrons.

Several Cd containers (cylindrical Cd-box, 2cm height x 1cm diameter) were made from a Cd foil (99.99% purity) with 1mm thickness (see Fig. 1). In spite of ENAA attractive characteristics its application faces some experimental difficulties: to achieve a considerable activity in the sample we need to apply longer irradiation times, inducing then a higher activity of the Cd container. This increases significantly the decay time needed before sample handling. Other problem is the container heating during irradiation, which may lead, in some cases, to destruction of the sample. We applied ENAA technique to various materials, in order to define the best experimental conditions: irradiation, decay and counting times. The samples analysed were: human urine, oyster tissue reference material, bovine liver reference material and fly ash on filter reference material. The results obtained were compared with those found by INAA analysis of the same material. Except for molybdenum, all the elements identified by INAA were also identified by ENAA. The increase in the peak area of some nuclides was evident as shown for arsenic in Fig. 2. In terms of ENAA of air particulate matter the long irradiation time needed lead to sample overheating and consequently loss of material. More investigation should be done.

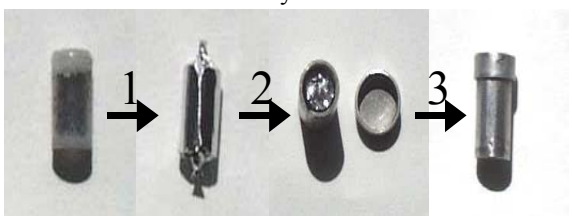
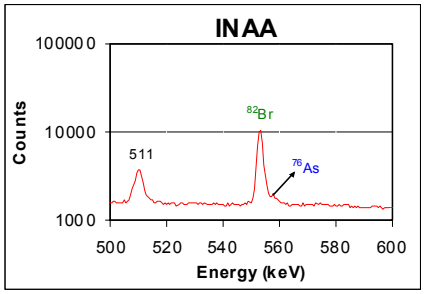


Fig. 1 Cadmium container used in ENAA.



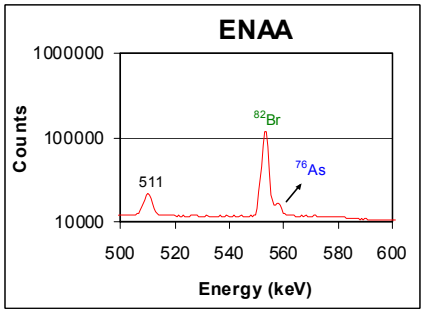


Fig. 2 Spectra taken for Human Urine.

Published, accepted or in press work

1. C.J. Costa, M.C. Freitas, Determination of Trace Elements in Human Urine using Neutron Activation Analysis, *1st International FESTEM Congress on Trace Elements and Minerals in Medicine and Biology*, Venice, Italy, May 16-19, 2001.

Experimental determination of RPI neutron flux and associated k_0 parameters

Carla Mustra, M. Carmo Freitas

Objectives

After a change on RPI core configuration, reactor parameters are modified and should be re-determined. Experimental values of parameters *f* (thermal to epithermal neutron flux ratio) and α (deviation of epithermal neutron spectrum from 1/E shape) were determined for irradiation sites 35, 56 and Be-N using "Cd-ratio for multimonitor" method". T_n (neutron temperature) and modified spectral index were obtained co-irradiating Lu with a "1/v" [$g(T_n)$=1] monitor.

Results

In actual irradiation sites, the epithermal neutron spectrum can be expected to deviate from the ideal 1/E shape. These deviating spectra in most cases can be approximated by:

$$\phi_e(E) = \phi_e \frac{1eV^{\alpha}}{E^{1+\alpha}}$$

where α is independent of energy and can be positive, negative or null, depending on the reactor configuration and location.

α determination

Parameter α is obtained from the slope (-α) of the graphical representation:

$$Y = \left\{ \log \frac{\overline{E}_{r,i}^{-\alpha}}{\left(F_{Cd,i} R_{Cd,i} - 1\right) Q_{0,}(\alpha) \frac{G_{e,i}}{G_{th,i}}} \right\} vs \log \overline{E}_{r,i}$$

where $F_{Cd,i}$ is the Cd-transmission factor for epithermal neutrons, $G_{e,i}$ is the correction factor for the epithermal neutron self-shielding, $G_{th,i}$ is the correction factor for the thermal neutron self-shielding, $Q_0(\alpha) = I_0(\alpha)/\sigma 0$ where I_0 is the neutron capture resonance integral, $R_{Cd,i}$ is the cadmium ratio.

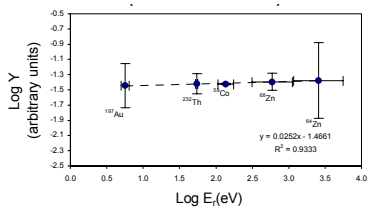


Fig.1 Experimental α and f determination in position Be-N of RPI.

***f* and $r(\alpha)\sqrt{T_n/T_0}$ determination**

The thermal-to-epithermal flux ratio, *f*, was also obtained graphically as the intercept with the ordinate [log (1/f)] of the plot presented before.

The application of parameter *f* on k_0 method formulated according Høgdahl convention, is restricted to (n,γ) reactions for which Westcott's g-factor equals unity, i.e. for which the cross section varies as σ(v)∝1/v (v-neutron velocity) in the thermal neutron region. T_o extend the method to non-1/v reactions, use was made of Westcott-formalism. This implies the experimental determination of the modified spectral index $r(\alpha)\sqrt{T_n/T_0}$.

Neutron temperature determination

The "absolute" T_n-determination is based on co-irradiating a Lu-monitor and a "1/v" monitor. Wires of 0.1% Au-Al and 0.1% Lu-Al with 1mm diameter, were irradiated in the three sites (35, 56, and Be-N). After measuring Wescott factor for lutetium $[g(T_n)]_{Lu}$ in each studied irradiation site, neutron temperature was obtained from tables $[g(T_n)]_{Lu}$ vs T_n. Table I presents the results of parameters α, f, T_n, and $r(\alpha)\sqrt{T_n/T_0}$ obtained for the irradiation sites 35, 56 and Be-N.

Table I Reactor parameters experimentally determined in irradiation sites 35, 56 and Be-N of RPI (ITN).

Irradiation Sites	***f* (*f*calc)**	**α**
Pos. 35	30.8±0.8 (40.2)	0.0369±0.0178
Pos. 56	98.19±1.9 (107.8)	0.0242±0.0096
Pos. Be-N	29.2±0.6 (35.7)	-0.0252±0.0108
Irradiation Sites	T_n **(ºC)**	$r(\alpha)\sqrt{T_n/T_0}$
Pos. 35	55	0.0284
Pos. 56	44	0.0097
Pos. Be-N	93	0.0303

Influence of γ Radiation and Dose Rate in Chemical Composition of Effluent Municipal Wastes

M.G. Ventura, L. Botelho[1], M.C. Freitas, C. Mustra

Objectives

Using INAA as analytical technique, we carried out a study in order to determine the influence of γ radiation and dose rate on the dynamic of cations between solid and liquid phase, present on wastewaters samples. It was used sewage from Beirolas (Portugal) wastewater Treatment Plant that is characterised as being predominantly domestic, although it may contain some industrial waste. The samples were obtained upstream to the treatment plant.

Results

Six sets of three wastewater samples of 40 ml each were filtered and two phases were obtained, a liquid and a solid phase (filtrate). Two of these sets contain samples irradiated in a Cobalt source with a dose rate of 0.78 kGy/h and the other two contain samples irradiated with a dose rate of 0.08 kGy/h, the remaining sets (two) are non-irradiated samples. Doses of 5 and 15 kGy were applied to each group of irradiated sets. The liquid phase was freeze-dried and a powder was obtained. All the solid phase samples were placed in high purity polyethylene boxes and irradiated. Gamma-spectra of the irradiated samples were collected using hyperpure germanium detectors of high resolution. Concentrations were determined using k_0-standardization method.

In Fig. 1 the evolution of Cr and As concentration as a function of γ irradiation is shown for high dose rate (HDR) and low dose rate (LDR) respectively. For Cr it can be seen that for the three doses applied the concentration in the solid phase, designated as filtrate, is always higher than that in the liquid phase. For As it can be noted that the element tends to dissolve.

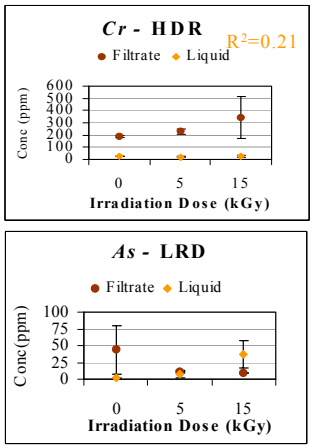


Fig. 1 As and Cr concentration evolution in function of irradiation dose at low and high dose rate, respectively.

Graphical results for Mn are presented in Figure 2. Mn presents higher concentrations in solid phase and as the irradiation dose increases, it is verified for both dose rates an increase in filtrate concentration and a simultaneous decrease in liquid phase concentration.

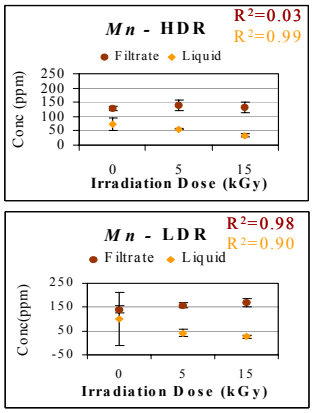


Fig. 2 Mn concentration evolution in function of irradiation dose at low and high dose rate, respectively.

Although some elements suggest a correlation between concentration and irradiation dose (see correlation factor), some set values showed a large dispersion. The large range of variance may be due to the formation of material agglomerates, in particular biological ones, which lead to a reduction in sample homogeneity. A more representative sampling, better sample homogenisation and previous combustion of organic matter could lead to better results.

Published, accepted or in press work

1. M.L. Botelho, M.C. Freitas, M.H. Gil, R. Zeinou, R. Ribeiro, S. Cordeiro, S. Cabo Verde, M.H. Casimiro, M.G. Ventura, C. Mustra, Treatment of Wastewater by Gamma Radiation – A Case Study, IAEA Regional Training Workshop – Radiation Treatment of Industrial and Municipal Wastewater, Istanbul, Turkey, November 18-22, 2001. Submitted to *Radiation Physics and Chemistry*.

[1] Radiation Technologies, Physics Sector