

## Reactor Sector





# Reactor

*José Gonçalves Marques*

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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 11% of the total irradiation time of the RPI in 2002.

The Reactor was the first sector to have an infusion of personnel that was essential to keep the RPI running and to reinforce its research activities. The average age of the RPI staff has decreased seven years in the last five years and it was still possible to put in contact two generations to minimize the loss of knowledge. However, this renewal has to continue and a new operators training course should start soon.

The staff involved in all aspects of the operation and use of the RPI presents its activities under the common headline of ***Operation and Exploitation of the Reactor, Dosimetry (RPI) and Reactor Calculations***. A significant number of research projects is going on, and funding through these projects increased 50% relatively to the previous year.

The ***Atmospheric Elemental Dispersion*** group was the main user of the RPI in 2002, accounting for 41% of the total irradiation time. The group uses the k0 INAA technique in the RPI, complemented by PIXE in the van de Graaff accelerator of the Physics Sector. The Atmospheric Elemental Dispersion group is

dedicated to cycling and impact of trace elements in the atmosphere. It addresses, specifically, the development and application of nuclear techniques, source apportionment and tracking in the atmosphere, chemical speciation, uptake and release of chemical elements in biomonitoring and monitoring, as well as health linkage through epidemiology studies. These objectives are approached through research, included mostly in MSc and PhD theses. The activities are financed by the industry (ValorSul urban waster incinerator) and by the Foundation for Science and Technology.

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

# Structure of the Sector and Technical staff

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## Research groups in the Reactor Sector

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

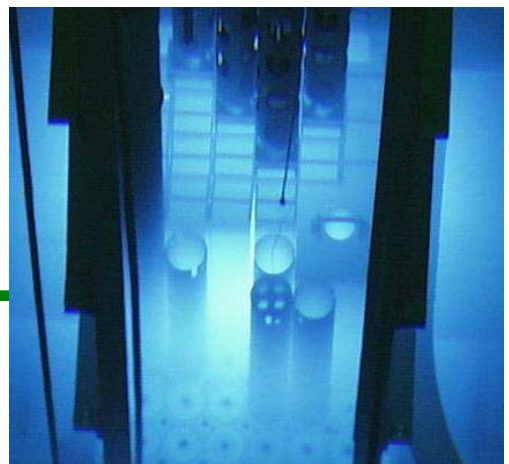
## Other Research Staff:

- J. OLIVEIRA (Co-ordinator Researcher)
- E. MARTINHO (Principal Researcher; until March)

## Administrative and Technical staff

- O. MENDES
- T. FERNANDES
- A. SILVA

## **Operation and Exploitation of the Reactor Dosimetry (RPI) and Reactor Calculations**





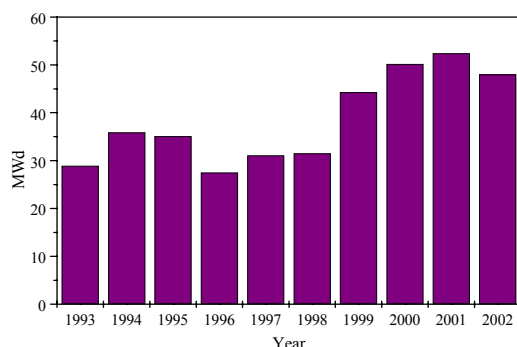
# Operation and Exploitation of the Reactor, Dosimetry (RPI) and Reactor Calculations

*J.G. Marques*

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

The programme for testing electronic circuits for cryogenic thermometry at LHC/CERN under fast neutron irradiation has continued during 2002. The irradiations were performed in a beam tube that was prepared for this effect. The same facility was used for the irradiation of citrus buds and rice seeds for IVIA (Spain) and for the irradiation of motion sensors for LIP/Lisbon. Other groups at CERN have also expressed their interest in using this facility next year.

The activity in the fields of dosimetry and BNCT has continued at a significant level and two PhD theses are being



finalized. The activity in dosimetry has also addressed the characterisation of several irradiation facilities, namely after the new core configuration N2-P1/7.

The operation of the reactor requires the calculation of parameters such as effective multiplication, control rod worth and safety parameters. The main drive of the work this year was comparison between calculated and measured values in the new core configuration. Calculations using low enriched fuel were also started, in order to prepare the core conversion to this type of fuel.

The activities using Monte Carlo codes have significantly increased this year, to areas such as the calculation of source terms, shielding and detector response. There is now a vigorous participation in the N\_TOF Collaboration at CERN and in a major European project on accelerator driven systems.

Concerning neutron scattering instruments, the Operation and Exploitation Group has introduced several improvements in the double axis spectrometer (DIDE) in collaboration with the Condensed Matter Physics Group. The floor in the detector's area was rectified, new power supply lines were installed and graphite crystals to improve the beam filtering were acquired and installed.

The installation of the Emission Channelling/Blocking Setup has been considerably delayed by the late delivery of the custom made position-sensitive detector. An effort will be made to recover this delay and have the facility in operation until next summer.

The main users of the reactor are described in the Table. Neutron activation analysis, to users in the Reactor and Chemistry, continues to be the largest activity. Isotope production has increased since last year, but its main users are still local research groups. The activity using the neutron scattering instruments has also increased and it is expected to further increase next year.

User	Area	Time (%)
ITN-RPI	NAA	40.7
	Dosimetry and BNCT	11.0
	Other (training, etc)	0.6
ITN-Chemistry	Isotope Production	13.1
	NAA	15.8
ITN-Physics	Beam Users	6.8
	Materials testing	0.7
Univ. Coimbra	Isotope Production	<0.1
CERN-LHC	Circuits Irradiation	10.8
IVIA	Irradiation of buds	0.4
LIP/Lisbon	Irradiation of sensors	<0.1

The figure indicates the integrated power produced by the RPI in the last 10 years. A clear increase is seen in the last 4 years reflecting an increase in its use. The integrated power for 2002 (until the first week of December) was 48.0 MWd, very close to the values of the last two years.

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

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## Research Team

### Researchers

- J.G. MARQUES, Auxiliary Researcher
- A.J.G. RAMALHO, Principal Researcher
- I.C. GONÇALVES, Auxiliary Researcher
- F. CARDEIRA, Auxiliary Researcher
- A. KLING, Auxiliary Researcher (90%)
- N.P. BARRADAS, Auxiliary Researcher (95%)
- A. FALCÃO, Principal Researcher
- A.R. RAMOS, Auxiliary Researcher
- P. VAZ, Principal Researcher

### Collaborators

- C. CHAUSSY (until August)
- J.-M. CHAUSSY (until August)

### Reactor Operators

- M.C. MARQUES
- R. CARVALHO
- J.A.M. RIBEIRO
- J.C. ROXO

### Funding (€)

Research Projects:	261 787
ITN:	72 947
Services:	10 188
<b>Total:</b>	<b>344 922</b>

### Publications

Journals:	2 and 2 in press
Proceedings:	2 in press
Conf. Communications:	3
Other publications:	8

- N. SERROTE

- V. PÁSCOA

- R. SANTOS

### Visiting Researchers

- A. VIEIRA, Co-ordinator Professor, ISEP

### Students

- M.J. PRATA, PhD Student, UL, FCT grant
- A. FERNANDES, PhD Student, UL, FCT grant
- J.A. SANTOS, ITN grant (until October)
- R. PATRÍCIO, PhD Student, IST/ITN

### Technical Personnel

- R. POMBO
- V. TOMÁS
- A. RODRIGUES
- J.S. SOUSA
- L.C. SANTOS
- J.L. SANTOS



## BNCT studies at RPI

I.C. Gonçalves, A.J.G. Ramalho, A.C. Fernandes, J. Santos, N. Oliveira<sup>1</sup>, M. Castro<sup>2</sup>, J. Rueff<sup>2</sup>

### 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 neutron capture reaction, are among the objectives of this project. The collaboration in a EU project to establish a Code of Practice for Dosimetry for BNCT in Europe, was pursued.

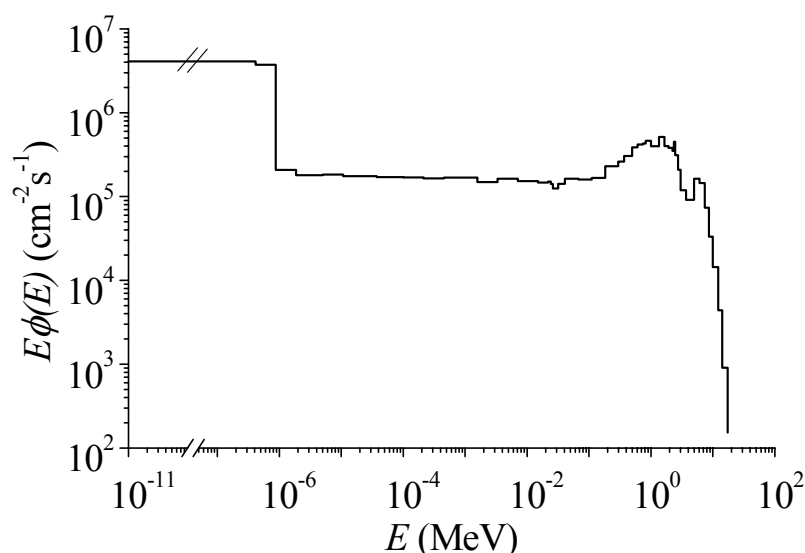
### Results

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.

Also the Co-60 source of DPRSN/ITN was used for gamma irradiations.

neutron capture reactions in the biological samples. The neutron dosimetry of the irradiations was performed with gold monitors.

The present study, using the cytokinesis-block micronucleus assay, reports on the differential effect of WM on the repair of the DNA damage induced by low LET (<sup>60</sup>Co  $\gamma$ -radiation) and high LET (boron neutron capture reaction,  $\alpha$  and Li particles) radiation in V79 Chinese hamster cells. Significant increases in the number of micronuclei per binucleated cell (MN/BN) as well as in the frequency of micronucleated binucleated cells (%MNB) were observed, in the presence of different concentrations of WM, for high LET radiation from the BNC reaction. The increases observed reached a maximum of about 2-fold in comparison to the respective controls. WM, however, had a more pronounced effect



The monitorisation of the irradiation conditions, mainly concerning the reproducibility of these conditions, is needed. The implemented procedure of gamma dosimetry in mixed radiation fields, with neutron insensitive dosimeters, was applied at the vertical access of the thermal column. TLD700 and a neutron insensitive ionisation chamber were used. The measured gamma dose rate was 400mGy/h. The MCNP code was used to calculate the dose due to the

on <sup>60</sup>Co  $\gamma$ -radiation-induced micronuclei, increasing the genotoxic damage from this radiation by about 3-4 fold. These results are in general in agreement with the concept that DSBs from high LET radiation may not be the more suitable substrate for these end-joining processes mediated by DNA-PK, yet they do not preclude a role for DNA-PK in high LET-induced damage repair.

### Published, accepted or in press work

1. N.G. Oliveira, M. Castro, A.S. Rodrigues, I.C. Gonçalves, O.M. Gil, A.P. Fernandes, J.M. Toscano Rico, J. Rueff, Wortmannin enhances the

induction of micronuclei by low and high let radiation, *Mutagenesis* (accepted).

<sup>1</sup>Fac. of Pharmacy, Univ.Lisbon,

<sup>2</sup>Dep. Genetics, New Univ.Lisbon.

# Improved Modelling of the Neutron Field at the Portuguese Research Reactor and its Experimental Validation

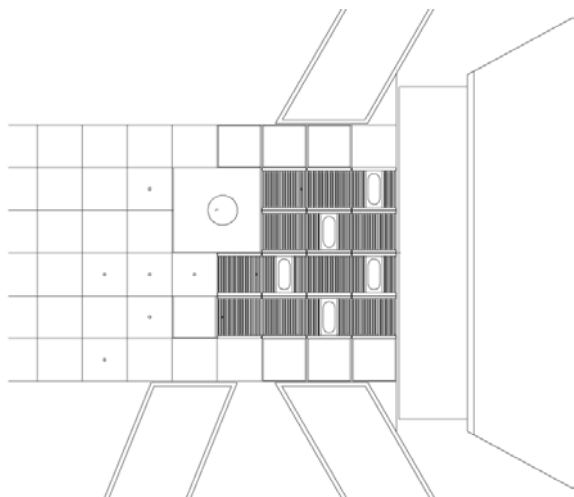
A.C. Fernandes, I.C. Gonçalves, N.P. Barradas, J. Santos

## Objectives

Simulation of the neutron field in the core and reflector regions of the Portuguese Research Reactor (RPI) with the Monte Carlo code MCNP-4C, using criticality and fixed-source calculations. Comparison of the MCNP results with those obtained with the deterministic codes WIMSD-5 and CITATION and with experimental results.

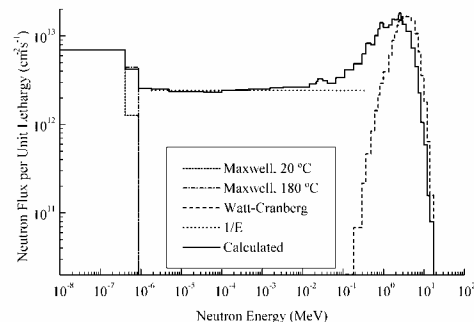
## Results

The RPI core model for MCNP was based on an accurate material and geometrical database. A cut view of the MCNP model is shown below.

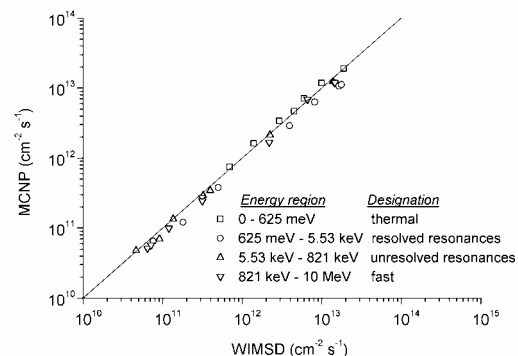


The value of the effective multiplication constant ( $k_{eff}$ ) was calculated in the criticality mode of MCNP. Compared to the WIMSD/CITATION results, there is a systematic deviation of  $\sim 0.016$ .

The neutron flux in the core and reflector regions, was calculated using two source models: the criticality mode and a fixed-source. Some discrepancies observed in the fuel plates, become progressively small farther away from the fuel, in the grid positions. Activation foil measurements were performed in the interplate spaces of the fuel assemblies and in some irradiation positions of the core grid. The activation detectors were Mn-Al 0.1%, Au-Al 0.1%, Lu-Al 1%, In, Ni and Al. In the figure below, a Maxwell, 1/E and Watt-Cranberg distributions are adjusted to the results.



The differences between the neutron fluxes calculated with MCNP and WIMSD (figure below) are  $(11.2 \pm 0.8) \%$ ,  $(28 \pm 2) \%$ ,  $(8.2 \pm 0.8) \%$  and  $(17 \pm 1) \%$ , respectively, for thermal, resolved resonances, unresolved resonances and fast energy regions.



The calculated response for selected activation reactions, at mid-height of the fuel assemblies, are shown in the table below, as well as the ratio of calculated to experimental values. An agreement better than 15 % was found for all cases.

Position	$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$		$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	
	C	C/E	C	C/E
N10, space 16	$9.29 \times 10^{-11}$	1.11	$1.85 \times 10^{-9}$	0.94
N7, space 2	$1.54 \times 10^{-10}$	1.05	$3.08 \times 10^{-9}$	0.94
Pos. 35	$2.62 \times 10^{-10}$	1.03	$2.57 \times 10^{-9}$	1.00
Pos. 36	$6.60 \times 10^{-11}$	0.89	$5.84 \times 10^{-10}$	0.90
Pos. 56	$4.69 \times 10^{-11}$	0.93	$3.92 \times 10^{-10}$	0.92
Pos. 17	$2.23 \times 10^{-11}$	1.01	$1.95 \times 10^{-10}$	1.00

## Published, accepted or in press work

1. A.C. Fernandes, I.C. Gonçalves, N.P. Barradas, A. J. Ramalho, Monte Carlo modelling of the Portuguese Research Reactor core and comparison with experimental measurements, *Nuclear Technology* (accepted).
2. A.C. Fernandes, I.C. Gonçalves, J. Santos, Improved modelling of the neutron field at the Portuguese Research Reactor and its experimental validation, *Proc. 11<sup>th</sup> Intl. Symp. Reactor Dosimetry, Brussels 2002* (accepted).

## Mixed-field dosimetry of a fast neutron beam at the Portuguese Research Reactor for the irradiation of electronic circuits

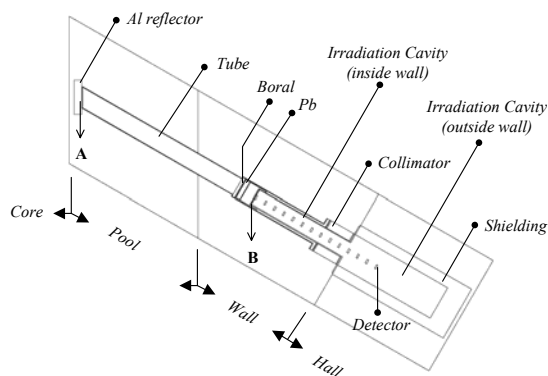
A.C. Fernandes, I.C. Gonçalves, J.G. Marques, J. Santos, A.J.G. Ramalho

### Objectives

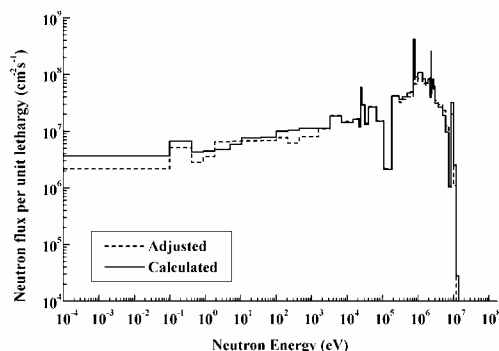
Simulation of the neutron and photon fields present at the Fast Neutron Beam of RPI with MCNP-4C and experimental measurements with activation foils, TLDs and ionisation chambers.

### Results

The Monte Carlo code MCNP-4C was used to calculate the various dose components of the beam. The MCNP model of the beam tube used for the simulations is shown in the figure below.



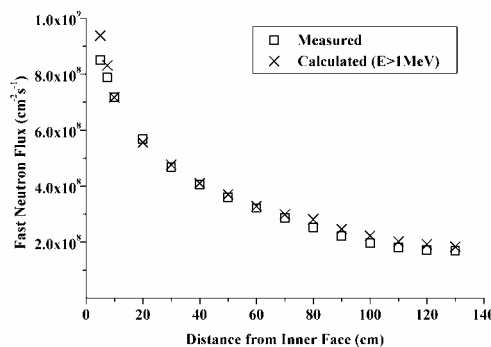
The neutron and photon sources used for the calculations were determined from a previous simulation of the core. The neutron energy spectrum and flux were measured with activation foils. Code LSLM-2 was used to unfold the neutron spectrum. The next figure shows the calculated and adjusted neutron energy spectra at point B.



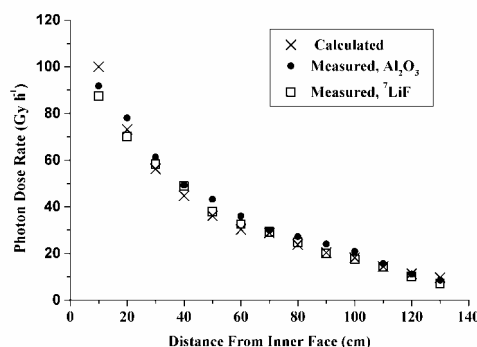
### Published, accepted or in press work

1. A.C. Fernandes, I.C. Gonçalves, J.G. Marques, J. Santos, A.J.G. Ramalho, Mixed-field dosimetry of a fast neutron beam at the Portuguese Research

The figure below shows the calculated and measured axial profiles of fast neutron flux.



Concerning the photon dose, a neutron-insensitive Mg(Ar) ionisation chamber was used. The photon dose profile (figure below) in the irradiation facility was determined with TLDs with low neutron sensitivity (TLD-700 and Al<sub>2</sub>O<sub>3</sub>:Mg,Y).



In general, there is a good agreement between calculations and measurements, although the model overestimates the thermal neutron component. Aluminum oxide TLDs were found to be promising for monitoring the photon dose in actual irradiations of circuits.

Reactor for the irradiation of electronic circuits – measurements and calculations *Proc. 11<sup>th</sup> Intl. Symp. Reactor Dosimetry*, Brussels 2002 (accepted).

## Fast Neutron Irradiation of Electronic Circuits for the LHC/CERN

J.G. Marques, I.C. Gonçalves, A.P. Fernandes, N.P. Barradas, F.M. Cardeira, A.J.G. Ramalho, J.A. Agapito<sup>1</sup>, F.J. Franco<sup>1</sup>, J. Lozano<sup>1</sup>, P. Gomes<sup>2</sup>, J. Casas<sup>2</sup>

### 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  $2 \times 10^{13}$  n/cm<sup>2</sup> 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  $5 \times 10^{13}$  n/cm<sup>2</sup> 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.

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 2002 as foreseen. Several power rectifiers, analog switches, digital-to-analog and analog-to-digital converters were tested under irradiation [1].

### 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.J.G. Ramalho, M.A. Rodriguez Ruiz, Data

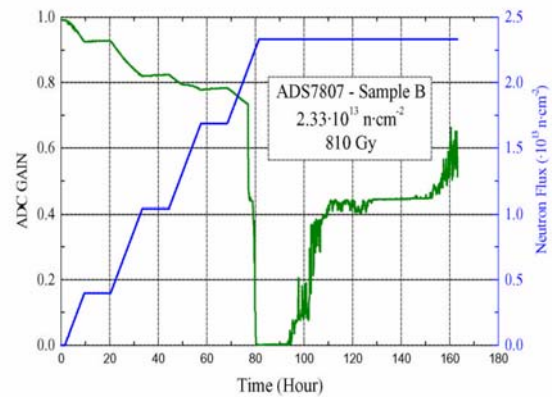


Fig 1. Gain of ADC circuit ADS7807 as function of the fast neutron fluence. The gain decreased abruptly after a fluence of  $1.6 \times 10^{13}$  n/cm<sup>2</sup>.

The irradiation of analog-to-digital converters was concentrated on the ADS7807 chip. This circuit is a parallel 16-bit, analog-to-digital converter built in CMOS technology by Burr-Brown. The offset and gain errors, effective number of bits and the internal reference voltage were measured during the irradiation. Half the converters used the internal reference voltage and the others an external one, previously qualified. Fig. 1 shows an example from an ADS7807 using the internal reference. The gain decreased abruptly after a fluence of  $1.6 \times 10^{13}$  n/cm<sup>2</sup>. The circuits using an external voltage reference had a better behavior.

This work will continue in 2003 with the testing of more components in statistically significant amounts. It is also foreseen that the testing of prototypes of the final circuit boards will start in 2003. A new irradiation device will be installed for this purpose.

Acquisition and Power Electronic for Cryogenic Instrumentation in LHC under neutron radiation, 8th Workshop on Electronics for LHC Experiments (extended abstract).

<sup>1</sup> Universidad Complutense de Madrid

<sup>2</sup> CERN, LHC/ACR Division

## Neutronics calculations for the RPI

N.P. Barradas, A.R. Ramos, J.G. Marques

### 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: neutron flux calculations, the core change to configuration N2-P1/7, and the effect of Xe poisoning. Furthermore, first calculations and developments of the calculation codes used envisaging the transition to < 20% enriched uranium forecast for 2006 were done.

### Results

The neutron fluxes were calculated in configuration N2-P1/7, and compared with measured values. The results, shown in Fig. 1, indicate a calculation error around 16%, which, considering a 12 year life-time of the Core 2 of the RPI, is good.

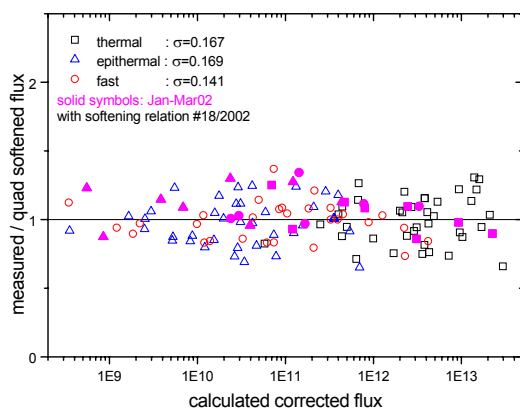


Fig.1. Dispersion of the ratio between experimental and calculated flux values. The data span 12 years. The solid symbols are for configuration N2-P1/7.

A burnup inventory of all fuel elements at the end of configuration N2-P1/6 was done, detailing the history of each fuel element and the corresponding burn-up at the end of each configuration N2-P1/1 to N2-P1/6. The  $^{235}\text{U}$  content of each element was also calculated.

### Published, accepted or in press work

1. J.G. Marques, N.P. Barradas, F.M. Cardeira, A.J.G. Ramalho, A.R. Ramos, Alteração da configuração do núcleo do RPI - Configuração N2-P1/7 - Relatório preliminar de execução, Janeiro 2002.
2. J.G. Marques, N.P. Barradas, F.M. Cardeira, A.P. Fernandes, I.C. Gonçalves, A.J.G. Ramalho, A.R. Ramos, Estudo da configuração N2-P1/7 do RPI - Configuração N2-P1/7, *Internal Report ITN/RPI-R-02/71*.

A new program, FLUXZONES, was written to calculate the average flux and macroscopic absorption and fission cross section for a given core. It was used to calculate those parameters in different configurations, that are used to calculate the Xe poisoning. Improved calculations for  $\Sigma_a$  were done, which led to a better agreement between the calculated

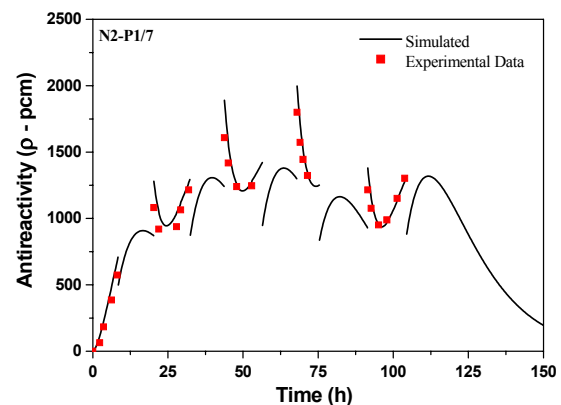


Fig. 2. Antireactivity caused by Xe poisoning in the N2-P1/7 configuration. Solid line corresponds to values calculated using a  $\Sigma_a$  varying with control rod position. Experimental data were taken during the first week of operation with the N2-P1/7 configuration.

and measured Xe antireactivity (see figure 2).

The code PREP1345, that prepares extensive parts of the CITATION input, has been previously developed at ITN. As it was written specifically for the Core 2 of RPI, it was now generalised. In particular, it was applied to the Core 1 of the RPI, which used <20% enriched uranium. Calculations were compared with data from 1961, resulting in good agreement. Internal ITN reports will be written in 2003. The driving force of these calculations was not an academic exercise, but to test the ability to calculate LEU cores.

## Monte Carlo Simulations for the Improvement of the RPI Health Physics Facilities

A. Kling

### Objectives

Keeping the Health Physics facilities at the RPI in an up-to-date state is a key issue in the operation of this research facility [1,2]. Monte Carlo (MC) simulations of radiation transport phenomena can be used to improve the radiological protection in many ways. Of special interest is the determination of detector responses to airborne radioactivity in the reactor hall and the stack exhaust of the reactor stack exhaust.

### Results

The airborne radioactivity in the RPI reactor hall is controlled in an air monitor with an open ionization chamber through which the air is pumped at a flow rate of about 100 litres per minute. The sensitivity of the system was so far only known for a few  $\beta$ -emitting isotopes ( $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{85}\text{Kr}$  etc.) from the technical specifications of the C/CAG 141 system. MC simulations using the MCNP code verified the response to these isotopes and enabled to determine the sensitivity to  $^{41}\text{Ar}$  and  $^{131}\text{I}$ , which are highly important in reactor operation. Figure 1 shows the results of the MC calculation of several isotopes and monoenergetic electrons together with known sensitivity values.

The values obtained are the basis for the revision of the alarm and warning limits for the airborne radioactivity at RPI.

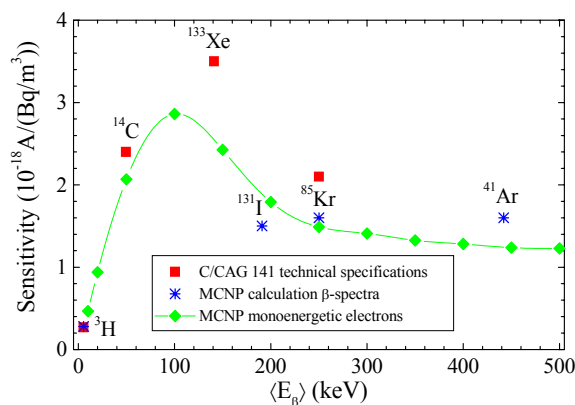


Fig. 1: Sensitivity of the air-monitoring system in the RPI reactor hall.

The gaseous radioactive effluents in the RPI stack exhaust are monitored by the AIRMON-91 system. The recalibration of the noble gas detector that controls the release of  $^{41}\text{Ar}$  requires an independent measurement of the air activity using a high purity germanium detector (HPGE) in a so-called Marinelli beaker geometry (see Figure 2). The response of the HPGE to a gaseous and uniform source of the 1.294 MeV gamma radiation of  $^{41}\text{Ar}$  has been computed using the MCNP. The optimum range for the dimensions of the Marinelli beaker has been determined varying its geometrical parameters. For the actual design parameters - a height of 200 mm and a radius of 100 mm - the expected photo peak efficiency is  $1.46 \pm 0.02\%$ . This efficiency value ensures that the  $^{41}\text{Ar}$  in the stack exhaust can be detected with the high accuracy required for the calibration.

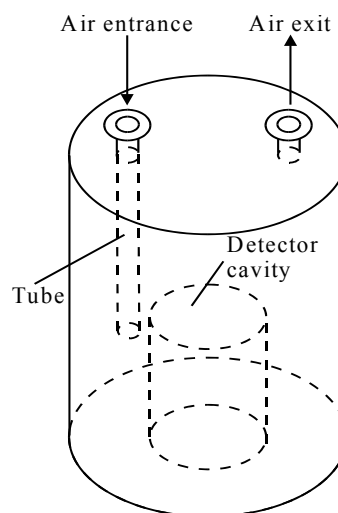


Fig. 2: Sketch of the Marinelli beaker to be used for the air-monitor calibration.

### Published, accepted or in press work

1. A. Kling, Radiological Control at the Portuguese Research Reactor (RPI) - Biennial Report 2000 - 2001, *Internal Report ITN/RPI-R-02/65*.
2. A. Kling, Radiological Control at the Portuguese Research Reactor (RPI) - Report January 1- June 30, 2002, *Internal Report ITN/RPI-R-02/68*.

## Studies on the Production of Lanthanide Radioisotopes at RPI

A. Kling, M. Neves

### Objectives

The evaluation of the capability of the RPI to produce radionuclides for the application is a fundamental step for the research on radiopharmaceuticals at ITN [1]. In this respect, radiolanthanides have been of increasing interest during the last years. Production of promethium by neutron capture in neodymium yields no-carrier-added isotopes, which are of special interest in medical applications. On the other hand, the influence of geometry and composition on the self-shielding in the case of irradiation of natural lutetium is of practical interest for the generation of  $^{177}\text{Lu}$  at RPI.

### Results

The irradiation of natural Nd with neutrons yields several Pm radioisotopes (from the decay of the Nd parent isotopes). Chemical separation of Nd and Pm after irradiation yields very high specific activity for the radioactive Pm. The most interesting isotope is  $^{149}\text{Pm}$  with a half-life of 53.08 hours. Figure 1 shows the calculated specific activities (with respect to the mass of Nd) for all Pm isotopes for irradiation cycles of 12 hours per day and a thermal neutron flux:  $\Phi = 1.6 \times 10^{13} \text{ n/cm}^2\cdot\text{s}$ , which are typical conditions for the RPI. The specific activity for the no-carrier-added  $^{149}\text{Pm}$  is 610 GBq/mg (16.4 Ci/mg). Although the activity arising from  $^{151}\text{Pm}$  can be reduced by a sufficient cooling time (ca. 10 days) the use of Nd enriched in  $^{148}\text{Nd}$  seems to be advisable for the production of this isotope.

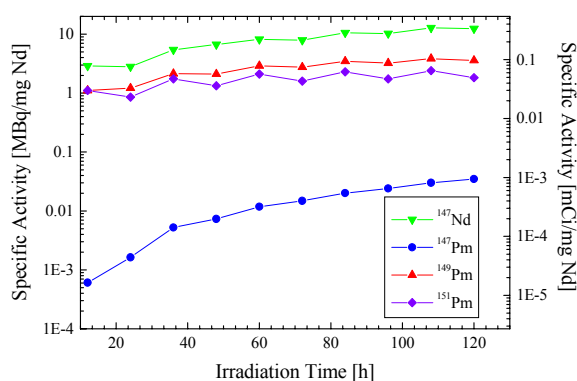


Fig. 1:  $^{147}\text{Nd}$ ,  $^{147}\text{Pm}$ ,  $^{149}\text{Pm}$ ,  $^{151}\text{Pm}$  specific activities vs. irradiation time for the irradiation of natural Nd.

$^{177}\text{Lu}$  is one of the promising  $\beta$ -emitting radioisotopes

of the lanthanide group for application in therapeutic radiopharmaceuticals. Although the use of Lu enriched in  $^{176}\text{Lu}$  has been shown to be a promising way to produce  $^{177}\text{Lu}$  with high specific activity and a minimum amount of the  $^{176\text{m}}\text{Lu}$  radio-contaminant [1] the irradiation of natural lutetium can be advantageous for basic research applications. Of special interest are the effects of self-shielding that limit the specific activity and their dependence on experimental parameters as geometry and composition. Therefore, a thorough study of these issues with special regard to the conditions at RPI has been performed [2]. Figure 2 shows as an example the dependence of the self-shielding of  $^{176}\text{Lu}$  on the sample thickness and the chemical form of Lu for the epithermal neutron energy region that exhibits strong resonances in the capture cross section.

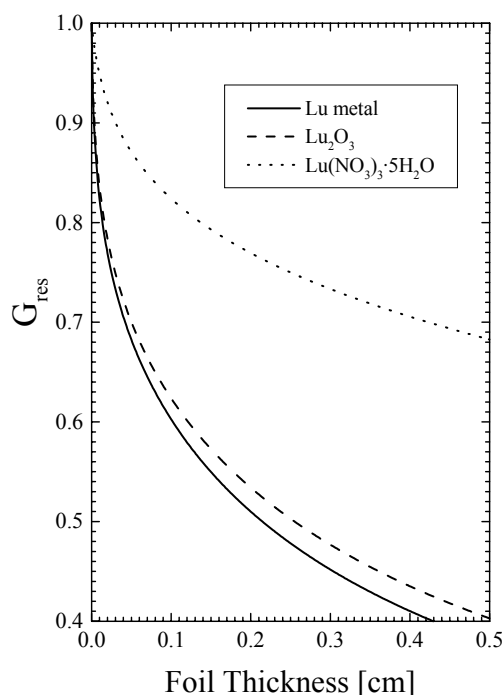


Fig. 2: Resonant self-shielding for  $^{176}\text{Lu}$  in foil-like geometry for different compounds containing natural lutetium.

### Published, accepted or in press work

1. M. Neves, A. Kling, R.M. Lambrecht, Radionuclide production for therapeutical radiopharmaceuticals, *Appl. Radiation and Isotopes* **57** (2002) 657-664.
2. A. Kling, Study on the Production of  $^{177}\text{Lu}$  by Irradiation of Natural Lutetium at the Portuguese Research Reactor (RPI) (Core Configuration N2-P1/7), *Internal Report ITN/RPI-R-02/67*.

## Progress of the Implementation of an Emission Channeling Setup at RPI

A. Kling, J. Marques

### Objectives

An emission channelling setup for studies on the structural properties of light elements as Li and B in single crystalline materials is under construction at the RPI. The angular distribution patterns arising from the channelling/blocking effect on charged particles produced in reactions with thermal and epithermal neutrons will be recorded by a position sensitive detector. After basic studies on the expected yields and shapes of the patterns the next step in the implementation of the experiment is the design of the reaction chamber and the readout electronics.

### Results

Figure 1 shows a cross section of the design for the reaction chamber in which the emission channelling experiments shall take place under ultra-high vacuum conditions. The main emphasis during the design has been laid on a structure that is mainly self-aligning in order to minimize the need for manual interventions during the studies.

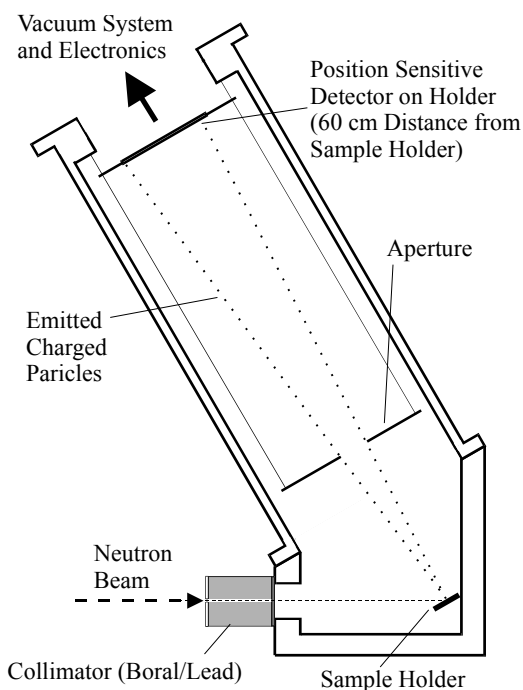


Fig. 1: Sketch of the irradiation chamber for the emission channelling setup

The double-sided position sensitive strip detector (Canberra PF-60CD-60CD-60\*60-300EB), mounted on a special holder, has been delivered during this year. This enabled us to advance with the design of the readout-electronics for the emission channelling setup. Figure 2 shows the schematics for the electronics to be used with the detector system. It will record the energy signal and the two position signals from the detector and enable the measurement of channelling patterns with 3600 pixels.

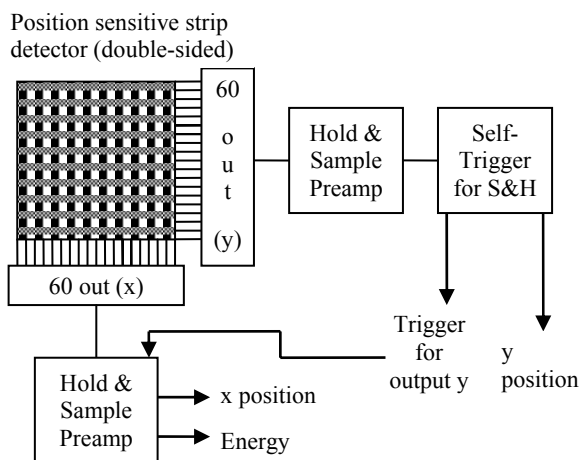


Fig. 2: Schematic of the readout electronics for the position sensitive detector.



## Improvements in the Radioprotection System of the RPI

Ch. Chaussy, A. Kling, A.R. Ramos, J.-M. Chaussy, J.G. Marques

### Objectives

Replace obsolete data recording units of the radioprotection system of the RPI by modern personal computers.

### Results

The current RPI radioprotection system was installed in the late eighties. It consists essentially of a set of fixed detectors from Merlin-Gerin installed at several points of the reactor hall and a stack monitor from the IAEA.

The Merlin-Gerin units are read by dedicated data processors with RS-232 outputs and analogue outputs. So far, only the analogue outputs were used to drive paper chart recorders. A new system developed at CEA was installed to read sequentially the data processors via the RS-232 interface into a dedicated PC-computer running MS-DOS. Another PC-computer running Windows NT acts as server and data archive. A dedicated program displays the values of up to four different detectors in real time on the computer monitor, chosen as the most important and representative in the reactor hall. Authorized computers within the ITN network can access the server at any time and read the current values of all fixed monitoring detectors in the reactor hall [1,2]. The new system has been in test operation since summer of 2002 without problems. In addition, a backup recording system was mounted, using a modern 8-channel paper chart recorder, which replaced eight obsolete thermal paper recorders. Figure 1 shows a view of the new integrated data monitoring and recording system.

The stack monitor, developed for the IAEA by the Seibersdorf laboratories, relied heavily on a 12-year-

old computer for data processing, archiving and periodic reporting. This computer was replaced in 2002 by a modern PC. It was necessary to adapt the original software, written in Basic for an 8088 processor at 8 MHz, to enable its proper operation on faster machines.



Fig. 1. The new integrated data monitoring and recording system.

Improvements were also made in data reporting. The locations within the reactor that require periodical monitoring by radioprotection personnel were revised, taking into account the new experiments installed.

### Published, accepted or in press work

1. C. Chaussy, Systeme de Centralization des Donnés de Radioprotection du RPI, *Internal Report ITN/RPI-R-02/69*.
2. J.M. Chaussy, Configuration Informatique du systeme de Centralization de donnés de Radioprotection du RPI, *Internal Report ITN/RPI-R-02/70*.

## Participation of ITN in the n-TOF experiment (PS213) at CERN

P. Vaz, C. Cruz<sup>1</sup>, I.F. Gonçalves<sup>1</sup>, J. Neves<sup>1</sup>, J. Salgado<sup>1</sup>, J.C. Soares<sup>1</sup>, R. Crespo<sup>2</sup>,  
L. Ferreira<sup>2</sup>, T. Peña<sup>2</sup>, L. Távora<sup>3</sup>, A. Melo<sup>4</sup>, L. Lourenço<sup>5</sup>, J. Lucas<sup>5</sup>

### Objectives

The n-TOF Collaboration, a consortium of 40 laboratories in Europe and U.S.A., has proposed an ambitious programme to perform high accuracy measurements of neutron cross-sections in the range from 1 eV to 250 MeV. An experimental programme (PS213) is being carried out since 2001 at the neutron time of flight (TOF) facility at CERN, using the CERN/PS accelerator complex. A single proton pulse of  $7 \cdot 10^{12}$  protons of 20 GeV impinges on a lead target every 2.4 seconds. After collimation, a neutron flux of the order of  $10^5$  neutrons/cm<sup>2</sup>/pulse is available for cross section measurements in the detectors station located 185 m downstream the target area.

These cross-section measurements are required in many emerging applications that require the use of high-intensity and medium-energy (in the hundreds of MeV) proton beams impinging on a thick target of an heavy element. These applications range from the design of innovative Accelerator Driven Systems (ADS) for incineration of nuclear waste and energy production, radioisotope production for medical and industrial applications and to many other subjects in Astrophysics, Nuclear Physics and Nuclear Technology. New or improved measurements of neutron cross-sections will also be very valuable for Radiation Shielding, Dosimetry and Monte Carlo Radiation Transport calculations.

### Results

This project deals with the following issues: i) radiation transport calculations using state-of-the-art Monte Carlo programs, ii) radiation detection and measurement techniques iii) particle detectors and associated electronics iv) high-precision measurements of neutron cross sections and v) physics analysis and nuclear data evaluation.

### Published, accepted or in press work

- 1.D. Cano-Ott, F. Calviño, G. Cortés, I.F. Gonçalves, E. Gonzalez, A. Poch, P. Vaz, Results of the Monte Carlo Simulation of the Neutron and Gamma Background in a Second Experimental Area for Fission Measurements at n-TOF, *CIEMAT report (2002)*.
2. I.F. Gonçalves, J. Salgado, L.M.N. Távora and P. Vaz, Towards the improvement of the neutron sensitivity of C6D6 detectors by using liquid scintillator material doped with heavy-Z elements, *to be submitted to NIM-B*.  
The n-TOF Collaboration, Measurement of the Neutron Capture Cross-Sections of <sup>232</sup>Th, <sup>231</sup>Pa,

During 2002, ITN researchers:

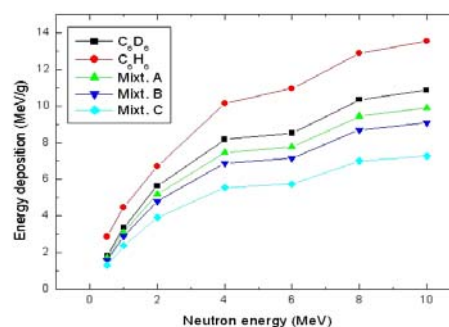
- Participated in the data taking shifts at CERN,
- Contributed to the Monte Carlo simulation studies to assess the new experimental conditions arising from future changes in the shielding and collimating systems of the TOF spectrometer,
- Simulated the response, both to neutrons and photons, of C<sub>6</sub>D<sub>6</sub> (deuterated benzene) liquid scintillator detectors and the properties of C<sub>6</sub>D<sub>6</sub> mixtures doped with high-Z elements,
- Promoted education and training initiatives in Nuclear Physics and related topics.

The following figure displays the simulated energy deposited by neutrons of different energies in a cylindrical volume filled with mixtures of C<sub>6</sub>D<sub>6</sub> scintillators doped with Tungsten:

Mixture A - C<sub>6</sub>H<sub>6</sub> (90%) + (C<sub>6</sub>H<sub>5</sub>)-W-(C<sub>6</sub>H<sub>5</sub>) (10%)

Mixture B - C<sub>6</sub>H<sub>6</sub> (80%) + (C<sub>6</sub>H<sub>5</sub>)-W-(C<sub>6</sub>H<sub>5</sub>) (20%)

Mixture C - C<sub>6</sub>H<sub>6</sub> (50%) + (C<sub>6</sub>H<sub>5</sub>)-W-(C<sub>6</sub>H<sub>5</sub>) (50%)



These computations were performed using the state-of-the-art Monte Carlo programs MCNPX, EGS4 and PENELOPE.

<sup>234</sup>U and <sup>236</sup>U, CERN/INTC-2002-013, proposal submitted to the CERN/INTC, April 2002.

The n-TOF Collaboration, Neutron Capture Measurements at the CERN n-TOF Facility for ADS Applications, *to appear in the Proceedings of the the CGS-11 Conference, Prague, 2002*.

The n-TOF Collaboration, Neutron Capture of Zr and La: Probing Neutron Exposure and Neutron Flux in Red Giant Stars, *proposal submitted to the CERN/INTC, September 2002*.

The n-TOF Collaboration CERN n-TOF Facility: Performance Report, CERN/INTC-O-011 (2002), *to be submitted to NIM-B*.

<sup>1</sup> ITN, Physics Department, <sup>2</sup> IST – (Technical University of Lisbon), Physics Department, <sup>3</sup> Centro de Instrumentação – Universidade de Coimbra, <sup>4</sup> CFNUL – Centro de Física Nuclear da Universidade de Lisboa, <sup>5</sup> ITN, Reactor Department, (project fellowships).

## Preliminary Design Studies of an eXperimental Accelerator Driven System (PDS-XADS)

P. Vaz, N.P. Barradas, R. Pires<sup>1</sup>, A.D. Sequeira<sup>2</sup>

### Objectives

The European Commission's Green Paper entitled "Towards a European Strategy for the Security of Energy Supply" clearly pointed out the importance of nuclear energy in Europe. With 145 operating reactors producing a total power of 125 GW<sub>e</sub>, the resulting energy generation of 850 TWh per year provides 35% of the electricity consumption of the European Union. The Green Paper also points out that the nuclear industry has mastery of the entire nuclear fuel cycle with the exception of waste management and for this reason, "focusing on waste management has to be continued".

Most of the hazard from the *spent fuel* from nuclear power plants stems from only a few chemical elements – plutonium, neptunium, americium, curium and some *long-lived fission products* (LLFP) such as iodine and technetium at concentration levels of grams per ton. At present approximately 2500 tons of spent fuel are produced annually in the EU, containing about 25 tons of plutonium and 3.5 tons of the "*minor actinides*" (MA) neptunium, americium, and curium and 3 tons of long lived fission products. These radioactive by-products, although present at relatively low concentrations in the spent fuel, are a hazard to life forms when released into the environment. As such, their disposal requires isolation from the biosphere in stable deep geological formations for long periods of time.

Accelerator Driven Systems (ADS) consist of a high-intensity proton accelerator of energies in the range 600-800 MeV impinging a target of an heavy element. The spallation reactions in the target material generate a very high neutron flux that feeds a surrounding sub-critical core system. The very high-flux of neutrons is required to sustain the *transmutation* processes by which the minor actinides and long-lived fission products can be "transformed" in nuclide species with a shorter lifetime, typically of hundreds of years (instead of dozens of thousands of years), and smaller radiotoxicity. Accelerator Driven Systems would have a great potential for nuclear waste transmutation and could be used to reduce the burden to underground repositories.

### Results

The PDS-XADS project is co-funded by the European Union under contract N° FIKW-CT-2001-00179. It is

### Published, accepted or in press work

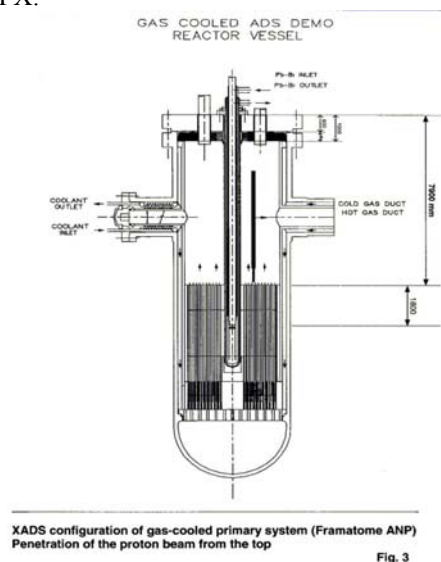
1. "Requirements for the XADS Accelerator & Technical Answers", *Deliverable D9 of the PDS-XADS project, Work Package 3*.

carried out by a consortium of 25 institutions (nuclear industry, universities and state laboratories) in European countries. Thw studies are organized around Work Packages (WP). The ITN team is directly involved, in the computational activities of:

**WP 4.3 – Target Unit design** – Spallation and neutronic calculations of the target unit (heat distributions, evaluation of the spallation and activation products, their energy and volume distributions). Work in cooperation with Framatome-ANP (WP leader), Ansaldo Nucleare (Italy), ENEA (Italy), FZK (Germany), SCK-CEN (Belgium), UPM (Spain), KTH (Sweden) and UMM (Poland).

**WP 3 – Accelerator studies** – Requirements for the operation of the XADS Accelerator (6 to 10 mA nominal proton beam current and 600 to 800 MeV energy) & the technical answers on issues such as stability, accidental situations and potential for other applications. Work in cooperation with FE-UCP (Faculdade de Engenharia da Universidade católica Portuguesa), CEA, CNRS and IN2P3 (France), INFN (Italy), IBA (Belgium), Univ. Frankfurt (Germany).

The following picture shows one of the possible configurations for the XADS in which the ITN team is involved. It consists of a gas-cooled Lead-Bismuth Eutectic target with a window separating the accelerator and the target. Studies and Monte Carlo simulations concerning this design are currently being performed using the state-of-the-art computer program MCNPX.

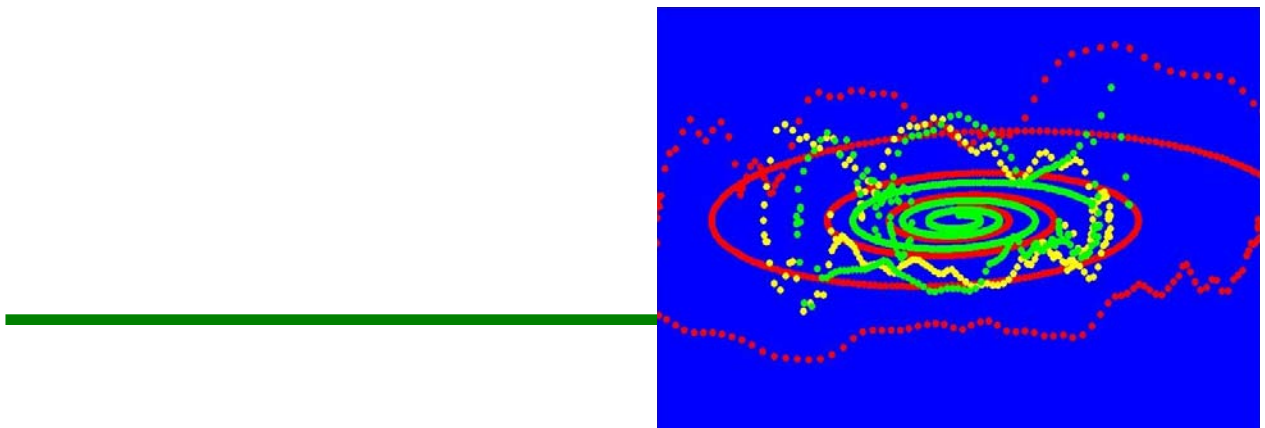


2. "Target Unit - Technical Specification and Target Unit Interfaces (LBE and Gas Cooled Concepts, Window and Windowless Options)", *Deliverable D70.1 of the PDS-XADS project, Work Package 4.3*

<sup>1</sup> FE-UCP - Faculdade de Engenharia, Universidade Católica Portuguesa, <sup>2</sup>ITN-Physics Department.



## Applied Dynamics





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# 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 also 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 courses in Portugal (Coimbra, Lisbon) to several post-graduation short courses 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, IPL, ENIDH) and abroad — France (Paris University), Ireland (Trinity College, Dublin), England (Southampton University) and Greece (University of Tessalonika).

Four research projects were pursued and two project were started at ADL during 2002, most of them funded by research contracts:

- Project “Simulation and identification of complex dynamic systems (ROTDYN)”, funded through the Sapiens programme;
- Project “Dynamical modelling of nonlinear systems (EXCITE)”, funded through the Sapiens programme;
- Project “Dynamical modelling of geological inclusions (GEOMODELS)”, funded through the Sapiens programme;
- Project “Optimisation of flexible-rotor balancing”, funded by EDP/CPPE.
- Project “Theoretical analysis of flow-structure coupled systems in nuclear facilities”, in collaboration with the French CEA;
- Project “Shape optimisation of vibratory and acoustic systems”, in collaboration with the LAM/Paris University.

The average time-scale of ADL projects is 2~3 years. One PhD thesis was recently concluded and three others are being prepared in connection with them. The main results of the research developed during 2002 are summarised in the following pages. For the second time, one of our research papers was awarded an international prize, from ASME.

# Applied Dynamics

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## Research Team

### Researchers

- J. ANTUNES, Principal researcher

### Collaborators

- L. HENRIQUE, Assistant professor (40%)<sup>1,2</sup>
- O. INÁCIO, Assistant professor (40%)<sup>1,2</sup>
- M. MOREIRA, Assistant professor (30%)<sup>1,3</sup>
- M. PAULINO, Assistant professor (20%)<sup>1,4</sup>
- R. SAMPAIO, Assistant professor (10%)<sup>5</sup>
- R. TABORDA, Assistant professor (10%)<sup>6</sup>

### Technical Personnel

- A. ANASTÁCIO

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<sup>1</sup> Doing PhD.

<sup>2</sup> IPP, Porto.

<sup>3</sup> IPS, Setúbal.

<sup>4</sup> IPL, Lisboa.

<sup>5</sup> ENIDH, Lisboa.

<sup>6</sup> FCL, Lisboa.

## Funding (€)

Research Projects: 27 481

**Total: 27 481**

## Publications

Books:	1
Journals:	4 and 4 in press
Proceedings:	6
Conf. Communications:	0
Other publications:	2
Theses: PhD	1



## Simulation and identification of complex dynamic systems (ROTDYN)

J. Antunes, M. Moreira<sup>1</sup>, H. Pina<sup>2</sup>, P. Piteau<sup>3</sup>

### Objectives

Development of new computational methods to dynamical problems of interest to industrial components. As a test problem, we work on rotor-flow coupled vibrations, which is a very important problem involving strong non-linear effects.

### Results

During 2002 our Galerkin/spectral theoretical formulation of immersed rotors to deal with orbital motions has been confronted with experimental work.

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 (120 pages), edited by WIT Press, currently in print.

Culminating several years of investigations, the PhD thesis of Dr. Miguel Moreira led to a significant number of international publications.

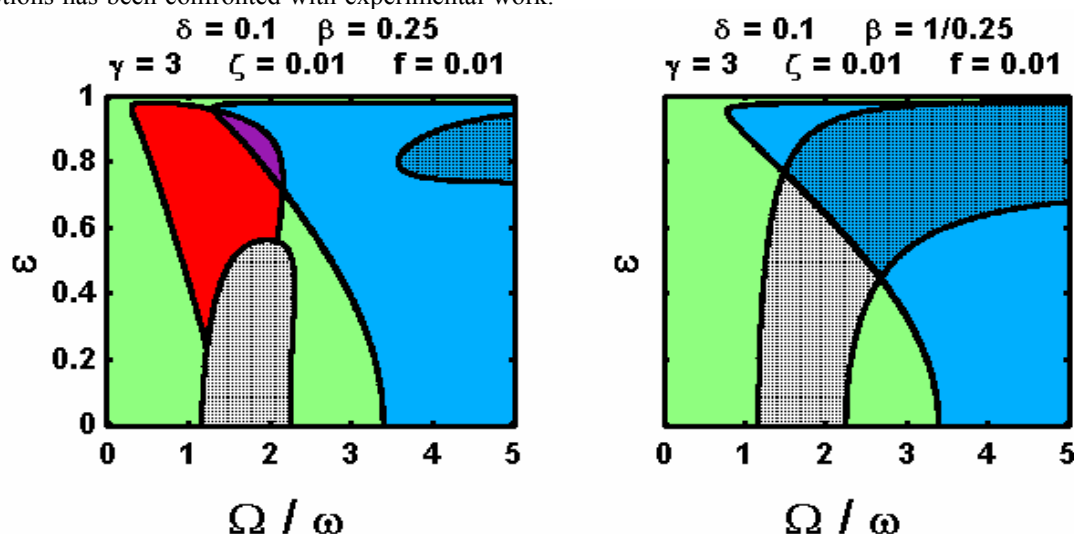


Fig. - Stability charts of an immersed rotor with asymmetric supports, as a function of the spinning velocity  $\Omega$  and of the rotor eccentricity  $\varepsilon$  inside the annular fluid gap. Each color represents a different dynamical regime: (Green = stable ; Other colors = instability of various combinations of forward and backward whirling modes).

### Published, accepted or in press work

1. J. Antunes, Dynamics of Rotor-Flow Coupled Systems, in Flow-Induced Vibrations, (P. Anagnostopoulos, Ed.), WIT Press, Southampton, UK (2002) (400 p.), ISBN 1-85312-644-6.
2. M. Moreira, Fluid-Structure Interaction in Linear and Nonlinear Rotor-Dynamics, PhD Thesis in Engineering Sciences, Instituto Superior Técnico, July 2002.
3. M. Moreira, J. Antunes, H. Pina, Nonlinear Analysis of the Orbital Motions of Immersed Rotors using a Spectral/Galerkin Approach, Communications on Nonlinear Science and Numerical Simulation, 7 (2002), 123-137.
4. M. Moreira, A. Tissot, J. Antunes, Experimental Validation of Theoretical Models for the Nonlinear Vibrations of Immersed Rotors, International Journal of Rotating Machinery, 8 (2002), 87-98.
5. M. Moreira, J. Antunes, H. Pina, A symbolic-numerical method for nonlinear rotordynamics under fluid confinement, International Journal of Nonlinear Science and Numerical Simulation, 3 (2002), 35-44.
6. M. Moreira, J. Antunes, H. Pina, An Improved Linear Model for Rotors Subject to Dissipative Annular Flows, Journal of Fluids and Structures, 2003 (in press).
7. M. Moreira, J. Antunes, H. Pina, Nonlinear Analysis of the Orbital Motions of Immersed Rotors using a Spectral/Galerkin Approach, Proc. of the ASME International Engineering Congress, July 2002, New Orleans, USA.

<sup>1</sup>Instituto Politécnico de Setúbal, IPS

<sup>2</sup>Instituto Superior Técnico, IST

<sup>3</sup>Commissariat à l'Energie Atomique, CEA/SEMT/DYN

## Dynamical modelling of nonlinear systems (EXCITE)

J. Antunes, O. Inácio<sup>1</sup>, L. Henrique<sup>1</sup>

### Objectives

EXCITE is an international co-operative effort to develop theoretical methods and numerical techniques to deal with strongly non-linear dynamical problems. 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 2002 we refined significantly our computational method for simulating the dynamics of bowed strings, through the inclusion of torsion modes

and also of the finite width – both are far from trivial developments. On the other hand, we also applied these computational techniques to bowed bars. In all cases, self-excited vibrations and bifurcation phenomena.

These techniques can be applied effectively to important friction-dominated problems, such as automotive brake devices. Experimental validation of our simulation results entrained significant difficulties, which delayed the main validation tests. However, a number of publications were produced from the theoretical results obtained until now.

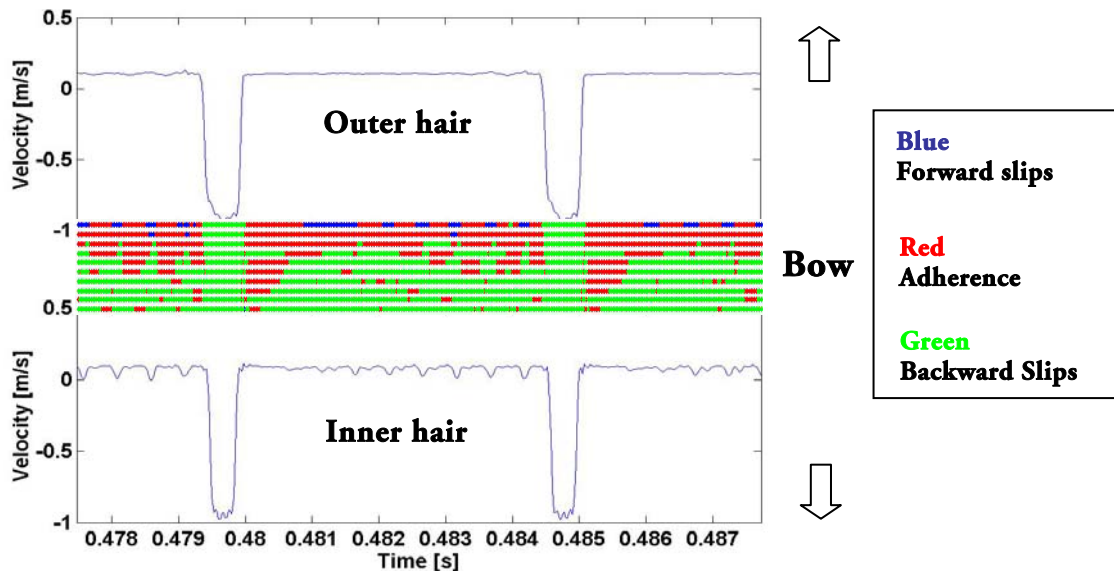


Fig.- Differential slip mechanism for a finite-width violin bow with complex slip-stick patterns.

### Published, accepted or in press work

1. O. Inácio, L. Henrique, J. Antunes, Simulation of the Oscillation Regimes of Bowed Bars: A Nonlinear Modal Approach, Communications in Nonlinear Science and Numerical Simulation, 2003 (in press).
2. O. Inácio, Largeur d'Archet et Régimes Dynamiques de la Corde Frottée, 6e Congrès Français d'Acoustique (CFA 2002), April 2002, Lille, France.
3. O. Inácio, J. Antunes, L. Henrique, Influence of torsion modes in bowed-string dynamics, Proc. of the 144th Meeting of the Acoustical Society of America & 3rd Iberoamerican Congress of Acoustics, December 2002, Cancun, México.

<sup>1</sup> Instituto Politécnico do Porto, IPP/ESMAE

## Dynamical modelling of geological inclusions (GEOMODELS)

F. Ornelas<sup>1</sup>, R. Taborda<sup>1</sup>, J. Antunes

### Objectives

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

The original project (Geomodels 1) has been significantly extended, as a new project (Geomodels 2). During 2002, the finite-element computation model previously developed for a Stokes-type flow has been refined, in order to accommodate also a matrix such

that shear stresses around the inclusion are almost null. This type of boundary condition also has great practical significance, leading to a quite different qualitative behaviour from the fully competent matrix case.

Extensive parametric computations have been performed, for both cases, and their correctness accessed against analytical solutions, for limiting cases.

Currently, our numerical solutions are being validated using the experimental work performed at FCL/LATEX. Several papers stemming from this work are being prepared.

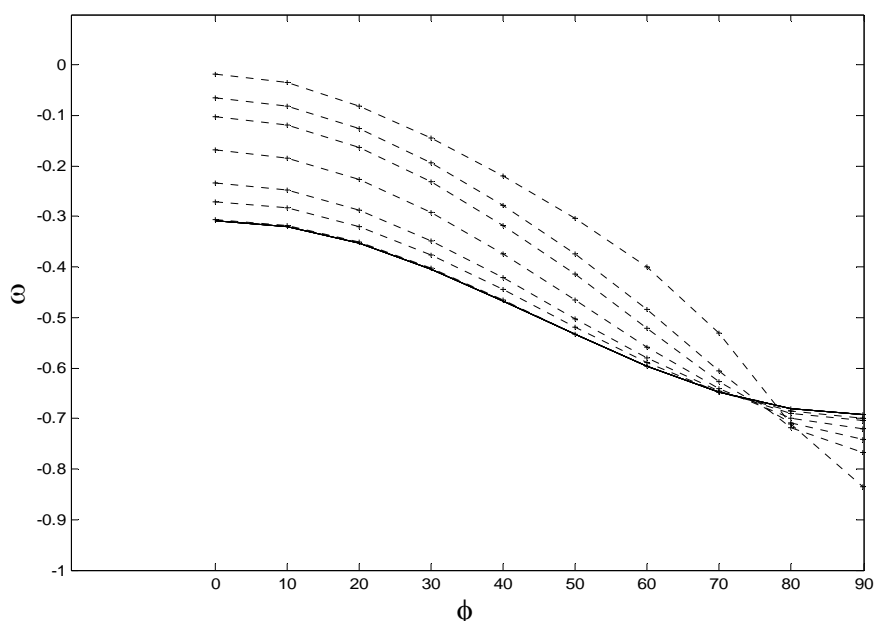


Fig.- Particle angular velocity ( $\omega$ ) as a function of the angle between particle major axis and the direction of simple shear ( $\phi$ ) for particles with an axial ratio of 1.5 and particle/channel confinement ratios between 1.6 and 10. The solid line represents the analytical solution for infinite channel width.

<sup>1</sup> Faculdade de Ciências de Lisboa, FCL/Structural Geology (LATEX)

## Optimisation of flexible-rotor balancing

J. Antunes, M. Paulino<sup>1</sup>, J. Soares<sup>2</sup>

### 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 have developed 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 pursued under contract for the Portuguese power supplier (EDP/CPPE). A PhD thesis is expected in connection with this project.

### Results

The validation tests scheduled for 2002, using a new and much improved test rig (SPIN/2), were delayed

significantly, due to experimental difficulties, as well as to the frequent unavailability of the test equipment, imperatively needed by CPPE for their maintenance field tests.

Therefore, in order to test the performance of our innovative approach for balancing, the main laboratory tests should be performed during the first months of 2003. However, we almost completed the development of our computer program for field balancing, which implements current methods as well as our new approach. It is expected that this balancing software will be extensively used by EDP/CPPE engineers in field balancing.

If the validating experiments produce the results expected, this project will lead to a number of publications.



Fig.- Impact test of improved experimental rig SPIN-2 for identification of the structural resonances.

<sup>1</sup> Instituto Politécnico de Lisboa, IPL/ISEL

<sup>2</sup> Electricidade de Portugal, EDP/CPPE

# Theoretical analysis of flow-structure coupled systems in nuclear facilities

J. Antunes, M. Moreira<sup>1</sup>, P. Piteau<sup>2</sup>, L. Borsoi<sup>2</sup>

## Objectives

Due to theoretical difficulties, a number of interesting problems connected to fluid-structure interaction and to flow-induced vibrations still lack adequate analytical solutions. However, such formulations are instrumental in order to achieve efficient time-domain numerical simulations of the nonlinearly-coupled systems when they are subjected to external excitations. Even more important, simplified formulations enable the prediction of self-excited vibrations under flow conditions, when the coupled system becomes linearly unstable.

This project addresses such issues, for specific problems motivated by components in nuclear power facilities. However, the results obtained may be extended to deal with problems arising in other physical and engineering systems.

## Results

The work carried out in this project during this year was focused on vibrations coupled with flows, mainly

axial in nature. Two specific problems were addressed:

(1) The *vibrations of spent nuclear fuel racks*, where the fluid couples the motions of several otherwise independent fuel racks. This problem is particularly significant under seismic excitation. An original simplified flow model has been formulated and the linearised problem was solved. Numerical simulations of a realistic system under random seismic excitation were produced [1].

(2) The *squeeze-film dynamics of vibrating structures subjected to axial flow*. This problem is significant for valves and many impacting components in several fields. A nonlinear flow formulation has been developed and analytically solved. Extensive numerical simulations were performed for both air-flows and water-flows. In particular, several kinds of instability phenomena were highlighted in our work [2].

This work was awarded an international prize for excellence, from the American Association of Mechanical Engineers (ASME).

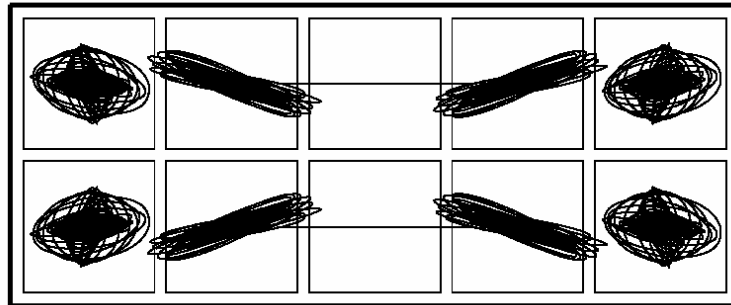


Fig. - Computed trajectories of 10 fuel racks immersed in a storage pool under fluid coupling when subjected to a seismic excitation.

## Published, accepted or in press work

1. M. Moreira, J. Antunes, A Simplified Linearized Model for the Flow-coupled Vibrations of Spent

Nuclear Fuel Racks, Journal of Fluids and Structures, 16 (2002), 971-987.

2. J. Antunes, P. Piteau, A Nonlinear Model for Squeeze-Film Dynamics Under Axial Flow, Journal of Fluids and Structures, 2003 (in press).

<sup>1</sup> Instituto Politécnico de Setúbal, IPS

<sup>2</sup> Commissariat à l'Energie Atomique, CEA/SEMT/DYN

## Shape optimisation of vibratory and acoustic systems

J. Antunes, L. Henrique<sup>1</sup>, J. S. Carvalho<sup>2</sup>, C. Besnainou<sup>3</sup>

### Objectives

Developing new methods for the optimisation of dynamical systems, focusing on the problem of shape optimisation, in order to obtain a target-set of resonance frequencies. As a sample problem, the tuning of percussion bars in marimba-like instruments was addressed. However, it is obvious that the techniques which are being developed can easily be used in many industrial problems.

### Results

Optimisation of dynamical systems is a field becoming each year more important. Indeed, economical reasons aggressively impose the replacement of costly and inefficient trial-and-error procedures for more formal and effective goal-directed computational approaches. Optimisation techniques, coupled with standard numerical techniques for the dynamical analysis of structural and acoustic systems are thus an extremely promising field.

Several optimisation methods have been implemented, including gradient-type local schemes, as well as a global optimisation algorithm – simulated

annealing – in order to avoid being trapped in sub-optimal local minima. Very interesting and satisfying results have been already obtained, however often at the expense of excessively long computing times [2]. On the other hand, extensive time-domain nonlinear numerical simulations of the impacted bars were performed, and the subjective qualities of the simulated sounds accessed [3].

Then, we developed a new optimisation approach, based on the description of the system shape in terms of a small number of orthogonal base-functions. This, coupled with an efficient local optimisation scheme, leads most often to satisfying results, with a two-orders of magnitude gain in computing time - when compared with the stochastic optimisation approach based on the physical degrees-of-freedom [1,4]. More recently, we started applying these methods to optimise the shape of acoustical systems – namely, instrument resonators.

This project is developed in collaboration with (and partly funded by) the Laboratoire d'Acoustique Musicale (LAM) of Paris University, in the framework of a larger network project.

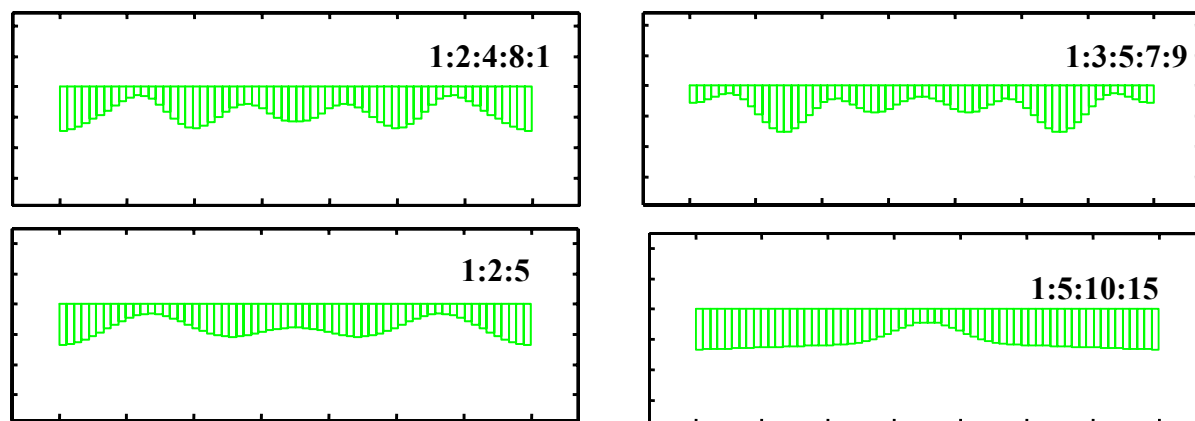


Fig.- Four examples of optimized profiles with bending modes at specified target frequency ratios.

### Published, accepted or in press work

1. L. Henrique, J. Antunes, Optimal Design and Physical Modelling of Mallet Percussion Instruments, Acta Acustica, 2003 (in press).
2. L. Henrique, J. Antunes, Conception Optimale des Instruments à Percussion : 1 – Stratégies d'Optimisation Géométrique, 6ème Conférence Française d'Acoustique (CFA 2002), April 2002, Lille, France.
3. J. Antunes, L. Henrique, Conception Optimale des

Instruments à Percussion : 2 – Synthèse Sonore par Modèle Physique, 6ème Conférence Française d'Acoustique (CFA 2002), April 2002, Lille, France.

4. L. Henrique, J. Antunes, J. S. Carvalho, Shape Optimization Techniques for Musical Instrument Design, Proc. of the 144th Meeting of the Acoustical Society of America & 3rd Iberoamerican Congress of Acoustics, December 2002, Cancun, México.

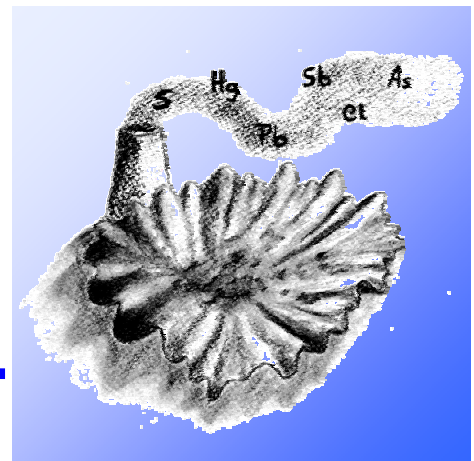
<sup>1</sup> Instituto Politécnico do Porto, IPP/ESMAE

<sup>2</sup> Universidade Nova de Lisboa, UNL/DCSH

<sup>3</sup> Université de Paris, LAM



## Atmospheric Elemental Dispersion







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# 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. The field is both a natural application of the potentialities of k0-INAA and PIXE techniques, which are the historical roots of the research 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 research unit activities include five main lines:

**1-Monitoring, Biomonitoring, Quality Control, Automation and Data Handling** aiming at characterising areas of Portugal 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 atmosphere, both locally and by long-range transport. In conjunction to this, data analysis methods 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, accuracy and precision studies on the nuclear techniques are being performed, both in biomonitoring and monitoring fields, aiming at better understanding differences found in the results for the same element and sample. Air particulate matter obtained by different air samplers is compared. So-called conventional analytical techniques are applied to complement the research unit's results. Automation and specific sampling equipment improvement research is carried out. This line is the major source of external financing.

**2-Atmospheric Trace Elements Effects Upon the Biosphere** includes 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 human being. Within this research program started health related problems. The

objective is to link biomonitoring to epidemiological studies, at local, regional and European scale.

**3-Chemical Element Speciation** aims at obtaining a better knowledge of the chemical state of the elements, its bioavailability towards the environment and its toxicity to the human being. Current biochemical, toxicological and physiological studies are focused not only on the overall occurrence of the element but also on their chemical forms. The specific item of chemical element speciation in atmospheric particles, biomonitors and total deposition is a full state of the art field being under development worldwide, almost at request of the Environmental Science Community. Presently the research unit proceeds with 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 and air particulate matter the state valence of the chemical element and the compound where it is included. This research is being done in close collaboration with groups outside ITN.

**4-Element Uptake Processes** describes the work to be performed in studies in which the relationships between elemental occurrences in lichens and in selected particulate matter size classes are investigated, thereby also considering elemental solubilities and extractabilities in both lichens and particulate matter. In addition, solubility is being studied in association to the elemental spatial distribution within the lichen organism. The underlying questions are related to the extent in which lichens may reflect the element contents of particulate matter, which may possibly be dominated by its soluble element concentration fractions.

**5-Training:** the research unit has a strong component in training. Four PhDs are ongoing (activities 1 and 3) and two PhDs are starting (activities 2 and 4). Two MScs in Nuclear Physics just started for a better support to the nuclear techniques application. Graduation theses are currently being completed.

**6-Services:** Analytical services are also provided under request.

# Atmospheric Elemental Dispersion

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## Research Team

### Researchers

- M.C. FREITAS, Principal Researcher (with “Agregação”)
- M.A. REIS, Auxiliary researcher

### Collaborators

- A.M.G. PACHECO, Auxiliary professor (15%)<sup>1</sup>
- H.Th. WOLTERBEEK, Senior researcher (10%)<sup>2</sup>

### Students

- S.M. ALMEIDA, PhD student, Aveiro Univ., PRAXIS grant
- C. HIPÓLITO, PhD student, Porto Univ., PRAXIS grant
- R. GODINHO, PhD student, Delft Tech. Univ., PRAXIS grant
- S. SARMENTO, PhD student, Delft Tech. Univ., PRAXIS grant
- A.P. MARQUES, PhD student, Delft Tech. Univ., ITN grant
- M.M. FARINHA, PhD student, Delft Tech. Univ., ITN grant
- M.G. VENTURA, MSc student, ITN grant

- O.R. OLIVEIRA, MSc student, ITN grant
- B. VIEIRA, Graduation student, Azores Univ.
- P. SOARES, Graduation student, Azores Univ.
- N. CORREIA, Graduation student, Azores Univ.
- P.C. CHAVES, Graduation student, Lisbon Univ.
- S. SIMÕES, BSc. student, ESTeSL
- S. VIGÁRIO, BSc. student, ESTeSL
- P. MATOS, Graduation student, ESA (50%)<sup>3</sup>

### Technical Personnel

- A. MACHADO, POCTI grant
- C. ROSAS, POCTI grant
- I. DIONÍSIO
- R. PINHEIRO (50%)<sup>3</sup>

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<sup>1</sup> CVRM-IST.

<sup>2</sup> IRI/Delft Tech. Univ..

<sup>3</sup> Shared with Physics Sector

## Funding (€)

Research Projects:	18 628
Industry Contracts:	93 014
Services:	150
<b>Total:</b>	<b>111 792</b>

## Publications

Books:	1
Journals:	4 and 7 in press
Proceedings:	7
Conf. Communications:	6
Other publications:	8
Reports:	7
Theses: LIC.	1
BSc	2

## Monitoring of Trace Elements in the Atmosphere Using Nuclear Analytical Techniques

S.M. Almeida, I. Dionísio, M.M. Farinha, M.C. Freitas, A.P. Marques, O.R. Oliveira, A.M.G. Pacheco<sup>1</sup>, R. Pinheiro, M.A. Reis, C.P. Rosas, M.G. Ventura

### Objectives

This program inherits work done by the members of the present DEA team since 1993 and combines both research and services to the community. The objectives are the direct monitoring of concentrations of trace metals in the atmosphere, a field where nuclear techniques can play an important role, and a field in which much support to the general society can be provided.

### Results

Within this program, and since 1999 services are being provided in the framework of the S. João da Talha Urban Residue Incinerator air quality monitoring program. Data on elemental contents in PM<sub>2.5</sub> and PM<sub>10-2.5</sub> are determined for: Pb (twice a week), Cr, Cu, Mn, Ni, As, Cd and Hg (once a week) and K, Sb, V and Zn (every two weeks). Air samples are collected during 24 hours using a Gent collector and analysed by INAA and PIXE techniques. Data are monthly reported. Results have shown that the EU limit value for Pb (500 ng/m<sup>3</sup>) was not over passed. A study carried out on the time evolution of Pb in PM<sub>10</sub> concentrations since 1994 in the Lisbon area. In July 2002, collaboration with the Institute of Meteorology (IM) was established aiming at the determination of the PM<sub>10</sub> composition out of the glass fibre filters used by the IM. Results of this program are presented in Table 1.

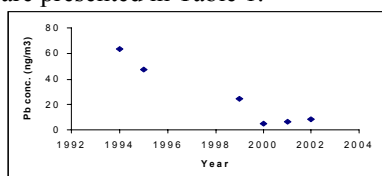


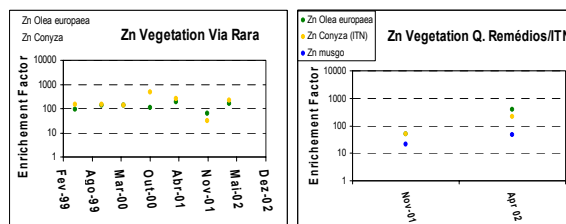
Fig. 1 – Pb PM<sub>10</sub> mass concentration evolution in Lisbon (in Ref. 1).

Table 1 – Content of trace elements in glass fibre filter

Metal	Known elements from literature (ng/cm <sup>2</sup> )	
	Literature	Determined
Cr	80	110
Fe	4000	1500
Zn	160000	160000
As	80	30
Sb	30	14
Other elements (ng/cm <sup>2</sup> )		
Na		425500
K		150000
Ca		50000
Sc		0.6
Co		0.9
Br		5
Ba		170000

ValorSul contract with biomonitors foresees a twice a year sampling, in Spring and in Autumn, in order to

monitor the trace element eventual increase due to the operation of S. João da Talha urban waste incinerator. Three biomonitors were chosen. This year 5 sampling points were proposed to Valorsul. Fig. 2 shows some



results for Zn an element associated with incinerators. Fig. 2 - Results for Zn in two sampling stations, Via Rara and Quinta dos Remédios/ITN.

A preliminary study has shown that bark from olive trees can be analysed in the same way as lichens or bryophytes, and for many of the same elements. Moreover, correlations between the concentrations of common tracers in bark and lichens are generally good, sometimes even significant at any level. Bark signals appear weaker than lichens', as expected, yet satisfactory to the extent that consistency is always much more an asset than magnitude proper (Fig. 3).

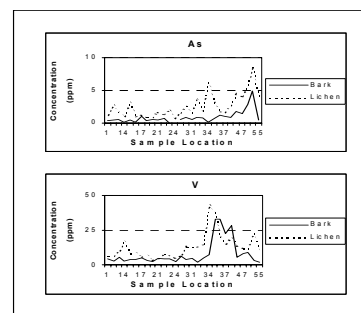


Fig. 3 – Concentrations for As and V for lichen and tree bark (in Ref. 2)

### Published, accepted or in press work

- M.C. Freitas, M.A. Reis, A.M.G. Pacheco, Recent trends in atmospheric lead levels at the urban-industrial district of metropolitan Lisbon, Portugal, through PIXE analysis of air-particulate matter, *Radiation Physics and Chemistry* (in press).
- A.M.G. Pacheco, L.I.C. Barros, M.C. Freitas, C. Hipólito, O.R. Oliveira, An evaluation of olive-tree bark for the biological monitoring of airborne trace-elements at ground level, *Environmental Pollution* 120 (2002) 79-86.

<sup>1</sup> CVRM-IST

## Quality Control of Trace Element Determination in Environmental Atmosphere Related Samples by Nuclear Analytical Techniques

S.M. Almeida, P.C. Chaves, M.C. Freitas, A. Machado, A.P. Marques, O.R. Oliveira, A.M.G. Pacheco<sup>1</sup>, R. Pinheiro, S. Simões, M.A. Reis, S. Vigário

### Objectives

The determination of atmospheric environmental parameters, from which chemical composition of aerosols and biomonitoring elemental content are but a part of the whole, requires precise and accurate values. The main reason is that data are compared 1) world wide, 2) to data from emission-based dispersion models, and are used in powerful statistical routines. This program aims then at data quality control.

### Results

Data quality work was directed to (i) elaboration of detailed procedures manuals for air particulate matter and biomonitor analysis and preparation; (ii) fast checking of samples history, including self controlled data handling routines with redundant steps to assure that no operator errors are propagated; (iii) the understanding of differences between PIXE and INAA values, which although in agreement considering the analytical uncertainties, differ often systematically.

#### Aerosols

Table 1. Contribution of blank filter Zn mass to Zn mass in filters (Ref. 1)

Zn conc(ng/m <sup>3</sup> )	INAA/PIXE	% contribution
< 10	3.5	17
>10 and < 50	1.5	7.3
> 50	1.3	2.7

A positive correlation (Ref. 1) was found between the % of blank correction and the (INAA/PIXE) ratios mean (Table 1). Also a clear positive correlation was found for 3 elements concerning their masses in the filter and the (INAA/PIXE) ratios mean. Accuracy studies were possible for INAA and not PIXE due to the unavailability of reference materials for this technique at the conditions used at our Institute. Using home-prepared and certified filters INAA accuracy could be ascertained as better than 6% for Zn, better than 5% for K, and better than 8% for Fe.

Data in Ref. 2 shows that the reproducibility varies from sample to sample and from element to element. An inverse relation between the elemental concentration and the agreement between parts of the same filter was detected, especially for PIXE analysis. Still, in general the results are reproducible to within 5-15%.

#### Biomonitors

Illustrative scatter plot for elements of interest from each origin is depicted in Figures 1 as an example (see Ref. 3). The results in Ref. 3 show that relative

deviations are well within commonly accepted limits of uncertainty for repeated experiments with a single technique, and that sign trends in such deviations are definitely not obvious. We also hypothesise that an improvement in chemical and physical homogeneity of PIXE samples may contribute to further abate that random variance.

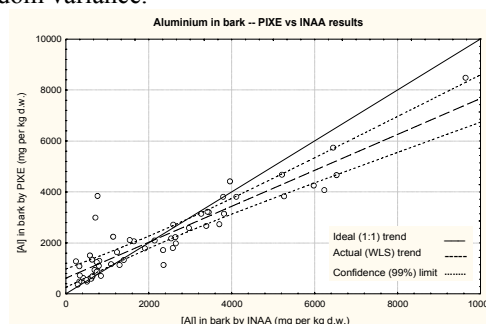


Fig. 1. Biplots of [Al] in olive-tree bark by INAA and PIXE.

Comparison of results of INAA and PIXE in lichens (see Zn data in Fig. 2) pointed out that the reproducibility also varies from sample to sample and element to element. The agreement is improved with an increasing (nr. PIXE replicates/nr. INAA replicates) ratio, and INAA/PIXE difference decreases with decreasing granulometry. .

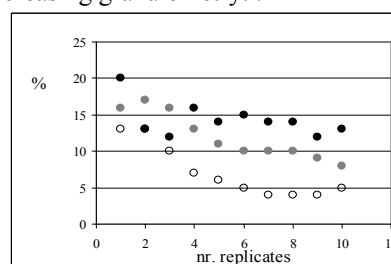


Fig. 2. black 125µm; grey 64µm; hollow 41µm

### Published, accepted or in press work

1. M.C. Freitas, S.M. Almeida, M.A. Reis, O.R. Oliveira; Monitoring trace elements by Nuclear Techniques in PM10 and PM2.5; *Nucl. Instr. Meth. in Phys. Res., part A* (in press).
2. S.M. Almeida, M.C. Freitas, M.A. Reis, Quality control of air particulate matter PM10 and PM2.5 by  $k_0$ -INAA, *J. Radioanal. Nucl. Chem.* (in press).
3. A.M.G. Pacheco, M.C. Freitas; M.A. Reis; Trace-element measurements in atmospheric biomonitors – a look at the relative performance of INAA and PIXE on olive tree bark, *Nucl. Instr. Meth. in Phys. Res., part A* (in press).

<sup>1</sup> CVRM-IST

## Biomonitors and Integration with Direct Aerosol Monitoring

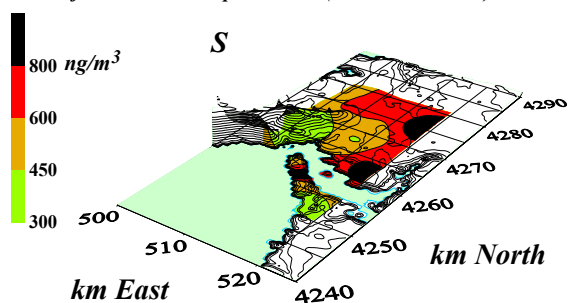
C. Costa, I. Dionísio, M.C. Freitas, A.P. Marques, O.R. Oliveira, A.M.G. Pacheco<sup>1</sup>, R. Pinheiro, M.A. Reis, M.G. Ventura

### Objectives

A very important issue in biomonitoring applications is the possibility of establishing the relation different biomonitors and between biomonitoring data and direct aerosol monitoring data. The study of the comparison between the information content in each of these types of data and the possibility of conversion from one into the other is the main aim of this program line.

### Results

*Reconstruction of mean of PM<sub>2.5</sub> airborne concentrations from lichen transplants data (UTM coordinates)*



*Reconstruction of standard deviation of PM<sub>2.5</sub> airborne concentrations from lichen transplants data (UTM coordinates)*

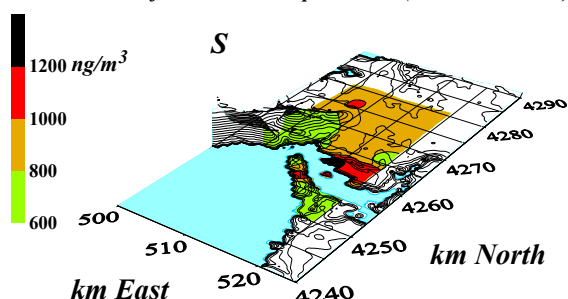


Fig.1 Mappings of nine months statistical distributions of concentrations of sulphur over the Sado Estuary study area, as inferred from lichen transplants data.

Within 2002 three types of work were carried out and/or published. The simplest of the approaches used

is based on the simple comparison of results obtained for the Sado Estuary region, based on wind differential biomonitoring (Ref. 2) and on direct monitoring.

Still discussing the data from the Sado Estuary experiment, and based in recently developed biomonitors calibration methods (Ref.1), it was possible to infer atmospheric concentrations out of lichen data. Mappings of the statistical distributions of atmospheric concentrations were thus constructed (Fig. 1) and the results obtained allow: (i) verifying how the selected aerosol sampling sites represent the regional values; (ii) locating sites where exposure may be expected to reach undesirable values; (iii) inferring probable sources based on the statistics mappings.

In the frame of biomonitors comparison, data from samples of the 1993 Portugal lichen survey, where olive tree bark was also sampled. In table 1 the comparison for a few crustal elements is presented.

In the conclusions of this work (Ref. 3) it becomes apparent that olive tree bark may be used as a substitute of lichen but, whenever regional mapping is the main issue, tree bark may even behave as less liable to signal inflation due to local sources than lichen samples.

Table 1. Distribution-free statistics and spurious-association probabilities for concentrations of crustal elements in *Olea europaea* bark and *Parmelia* spp. thalli across the study area (pooled sectors; number of valid cases) (in Ref. 3).

		Kendall $R_k$	$p$ -level	Correlation
BARK vs [Al] <sub>LICH</sub>	(n = 57)	0.410	0.000(007)	Excellent
BARK vs [Fe] <sub>LICH</sub>	(n =	0.278	0.004	Very high
BARK vs [Rb] <sub>LICH</sub>	(n = 52)	0.611	0.000(000)	Excellent
BARK vs [Se] <sub>LICH</sub>	(n =	0.248	0.009	Very high
BARK vs [Ti] <sub>LICH</sub>	(n =	0.109	0.356	None

Nominal qualifiers for the correlation status:  $p < 0.001$ : excellent;  $0.001 \leq p < 0.01$ : very high;  $0.01 \leq p < 0.05$ : high;  $0.05 \leq p < 0.10$ : fair;  $0.10 \leq p$ : faint to none.

### Published, accepted or in press work

1. M.A. Reis, L.C. Alves, M.C. Freitas, B. Van Os, J. De Goeij, H.Th. Wolterbeek, Calibration of lichen transplants considering faint memory effects, *Environmental Pollution* 120 (2002) 87-95.
2. C.J. Costa, A.P. Marques, M.C. Freitas, M.A. Reis, O.R. Oliveira, A Comparative Study for Results obtained using Biomonitors and PM<sub>10</sub> Collectors

in Sado Estuary, *Environmental Pollution* 120 (2002) 97-106.

3. A.M.G. Pacheco, M.C. Freitas; M.G. Ventura, Relative enrichment of trace elements in atmospheric biomonitors – INAA results on tree bark and lichen thalli, *Proceedings of the International Conference on Applications of high precision - Atomic and nuclear methods*, Hipan 2002, Neptun, Romania September 2-6, 2002.

<sup>1</sup> CVRM-IST

## Source Identification and Tracking

S.M. Almeida, M.C. Freitas, A.P. Marques, O.R. Oliveira, C.A. Pio<sup>1</sup>, M.A. Reis, M.A. Trancoso<sup>2</sup>

### Objectives

Source identification and tracking is one of the major fields of application of Nuclear Analytical Techniques (NATs) in atmospheric environmental studies. This is therefore a horizontal program within the group, and aims at the development and application of methods for the identification of emission sources signatures and location. Out of the whole set of methods available the group fine tunes its selection upon those which make best use of the multielemental characteristics of the data gathered by the group.

### Results

Within the framework of methods application two data sets were used: (i) a data set obtained from a 2001, one year atmospheric suspended particulate matter (PM) sampling campaign, carried at ITN campus, which used both a low and a high volume samplers working simultaneously during 24 hours periods twice a week; (ii) a data set obtained from a lichen transplants experiment carried out at the Sado Estuary.

Data obtained in database (i) originates both from: INAA and PIXE analysis of low volume samplers polycarbonate Nuclepore membrane filters, carried out at ITN; and from Chromatography, Atomic Absorption Spectroscopy, Potentiometry; and UV Spectroscopy analysis of high volume sampler filters carried out at INETI.

Factor analysis was applied to elemental data from Nuclepore filters and three main sources of PM were identified, namely: data soil (PC1), sea spray (PC3) and industrial (PC2, PC5 and PC6) (Table I).

Table I: Factor analysis applied to PM2.5

	PC1	PC2	PC3	PC4	PC5	PC6
PM2.5	0.25	0.50			0.38	0.51
Na		-0.31	-0.91			
Al	0.97					
Si	0.99					
S						0.85
Cl			-0.80			-0.32
K	0.55	0.50				0.29
Ca	0.83					
Sc	0.99					
Ti	0.94					
V					0.94	
Mn	0.81	0.34				
Fe	0.92	0.27				
Co	0.54				0.65	
Ni					0.93	
Cu		0.80				
Zn		0.78				-0.25
As		0.77			0.41	
Se			-0.99			
Br			-0.81			0.33
Sb		0.72			0.26	0.41
La	0.98					
Ce	0.95					
Sm	0.99					
Hg			-0.99			
Pb		0.51			0.57	
Eigenvalues	9.53	3.60	2.24	2.09	3.08	1.69
Per cent of total variance explained	36.7	13.8	8.6	8.1	11.8	6.5

<sup>1</sup> Univ. Aveiro

<sup>2</sup> INETI

Sources were also inferred based on statistics mappings with the lichen transplants data set. These results can be compared to Monte Carlo Target Transform Factor Analysis (MCTTFA).

MCTTFA analysis was performed in various ways. In Fig. 1, mappings of factor intensity for a sea salt source and a physiological factor are presented.

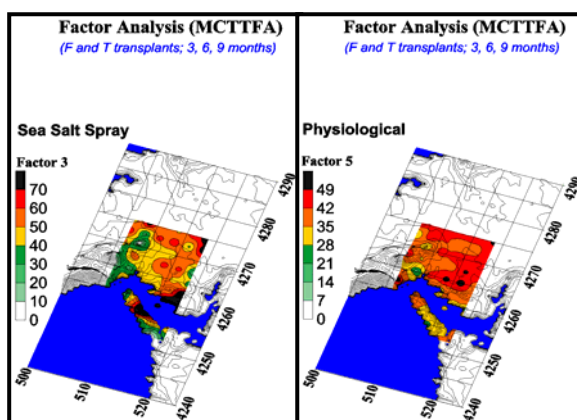


Fig. 1 – SURFER mapping for the factor values obtained by MCTTFA for two identified factors in Sado estuary study.

In what relates the development of methods, the full theoretical background of a new robust method that is being developed by the team for some years now was finally well established in 2002. This method, named ECRA, will allow in the future (after some software development and implementation), the determination of unknown sources, including sinks and sink-sources processes, and using very few samples. In a communication sent to press, it is shown that the use of 10 samples with data on 10 elements will allow the determination of as much as 50 sources and sinks.

### Published, accepted or in press work

1. S.M. Almeida, M.C. Freitas, M.A. Reis, C.A. Pio, Air particulate matter characterization and source identification in the North of Lisbon area, Proceedings of the conference: 6<sup>th</sup> International Aerosol Conference, Taipei, 8-13<sup>th</sup> September, 2002.
2. M.C. Freitas, M.M. Farinha, C. Costa, A.M.G. Pacheco, Measurement of Selenium in PM2.5 by INAA at a Urban Industrialised area of Portugal, Sustainable Development of Energy, Water and Environment Systems, Dubrovnik, Croatia, June 2-7, 2002.



## Studies and Development on Transplants Methodology

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### Objectives

For several years now, the Atmospheric Elemental Dispersion Group has been studying methods aiming at the improvement of the potentialities of biomonitoring using transplants and the increased understanding upon the behaviour and response of transplants to the surrounding environment. This program is being carried out in strong cooperation with the Delft Technology University, and more recently with the University of Porto (both LAQUIPAI and CIIMAR research centres) and CVRM-IST.

### Results

In the past months, a preliminary study has been running related to Biocal project. With this project, we intend to calibrate *in situ* monitors (lichens) with alternative *in situ* biomonitor (tree bark) and transplants (resins and lichens). The main objective of this preliminary assay is to determine the response of several monitors systems to element absorption.

At Sado estuary a case study enhancement by wind direction differential exposure of biomonitor was verified and cell membrane damage in lichen *Parmelia sulcata* was studied.

In the first stage of the Biocal project, different types of biomonitor (lichen and lichen biomass transplants) and monitor (cationic resin) were exposed in Porto city for several periods of time. The monitors systems consisted in the monitor itself attached to a plastic support with linen. For control of the experience two systems were exposed in the same period of time: one consisted in an inert material (cellulose acetate) and the other in linen. The concentrations obtained with the exposed samples were compared with a background. The analysed samples of lichen transplants (*Parmelia caperata*) indicated a positive response for several elements. Figure 1 is representative of that evolution.

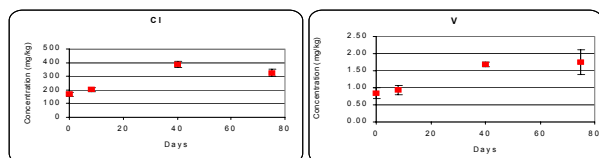


Fig.1 Evolution of the contents of elements in exposed lichens transplants (*Parmelia caperata*) determined by INAA

A decrease of concentration was observed in the physiological elements (K, Mn and Rb), indicating serious damages in the cellular structure of these

lichens. Another interesting response was obtained with the analysis of the inert material. Although not expected, there was some positive response with the cellulose acetate. Fig 2 is representative of that evolution. The figure clearly indicates a positive uptake of the elements by the inert material (the evolution is similar to every analysed elements).

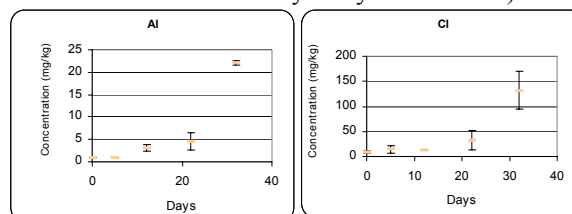
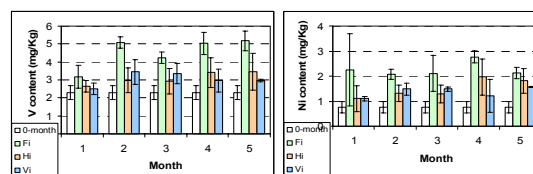


Fig.2 Evolution of the concentration of elements in exposed cellulose acetate determined by INAA

Concerning the Sado case study, lichen transplants were exposed in 3 different set-ups at ITN campus, allowing i) free elemental influx, ii) horizontal influx only and iii) vertical influx only. Samples were analysed by INAA and PIXE. Fig.3 shows some



results.

Fig. 3 V and Ni results evolution during 5 months exposure for the 3 different set-ups.

Electrical conductivity was measured to access cell membrane damage in above reported *Parmelia sulcata* transplants (see Fig. 4).

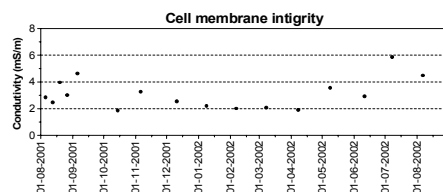


Fig. 4 – Results obtained for conductivity measurements.

### Published, accepted or in press work

1. H. Th. Wolterbeek, J. Garty, M.A. Reis, M.C. Freitas, Biomonitoring in use: lichens and metal air pollution; in: Bioindicators and biomonitoring, Chapter 11 (pp. 377-419), B.A. Markert, A.M. Breure, H.G. Zechmeier, editors, 2002 Elsevier Science B.V.

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## Instrumentation for Aerosol Characterization using Nuclear Analytical Techniques

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### Objectives

This project aims at the improvement of existing instrumentation for both aerosol sampling and characterization. Sampling instrumentation, as well as instrumentation associated to the analytical techniques themselves, is therefore considered under this project.

### Results

Under this wide scope the following issues were addressed during 2002: 1- increase of the sampling equipment URSA and the ITN-PM10 Separator reliability; 2- improvements on the PIXE analysis chamber.

1- (i) Some heating problems were detected and solved: undesirable gas heating due to vacuum pump functioning was solved applying a copper-brass high surface thermal dissipater and a fan at the pump's exit. Air temperature entering the flux meter and the volume integrator is thus lowered to ambient values. Errors due to diaphragm pump heating were therefore eliminated. Re-dimensioning thermal dissipaters and repositioning components were also carried out to reduce electronic based heating.

Comparison between the URSA unit using the ITN-PM10 Separator (without the heating unit working), and a Gent unit, in real working conditions, provided the results presented in Table 1. These results are compared to a subset of 11 results extracted from a 104 samples experiment where two Gent systems were compared. (ii) Some strange results in sampling parameters were found which were attributed to the fact that under certain atmospheric conditions water vapour might condense inside the aerosol separator, altering the behaviour of filters and raising questions over the reliability of the aerosol collection process. Study of gas-liquid transition of atmospheric water vapour lead us to the development and construction of a heat ring inside the ITN PM10-Separator. The ring is expected to heat the air being filtered, therefore decreasing the possibility of water condensation.



Fig.1: Partial view of the meteorological station at ITN campus: thermo-hygrometer, rain collector and radio-transmitters.

(iii) Physical and chemical properties of aerosol particles are also dependent of atmospheric parameters. A low cost meteorological station (Fig. 1) was thus implemented at ITN campus at the aerosol collection test site. The station monitors air temperature, relative humidity, barometric pressure, rainfall, wind speed and direction in real-time.

Table 1. Student t-test for identity between the URSA and the Gent results. 11 samples were used for this comparison. Comparison was made using a pair wise, double-tailed approach. G vs G (104) presents the results for the 104 samples experiment.

	t value	p value	null hypothesis identical (0.05)
<b>PM2.5</b>			
URSA vs Gent	1.27	0.23	identical
Gent vs Gent	-3.68	0.004	NOT identical
G vs G (104)	-0.92	0.36	identical
<b>PM10-2.5</b>			
URSA vs Gent	-2.32	0.04	NOT identical
Gent vs Gent	-3.38	0.007	NOT identical
G vs G (104)	-1.89	0.06	identical
<b>PM10</b>			
URSA vs Gent	-1.24	0.24	identical
Gent vs Gent	-4.52	0.001	NOT identical
G vs G (104)	-1.68	0.09	identical

In what refers to item 2, the following developments were made on the PIXE chamber:

(i) a new, more efficient, electron gun was developed; (ii) a new chamber's back was built which includes a window for easy on-the-run checking of beam quality; (iii) a ~400Hz low voltage noise signal originated at the stepper-motor commuted power source was eliminated by powering the system with a lead-acid 12Vdc battery; (iv) finally, a CdTe detector which is to function simultaneously with the Si-Li detector, reducing by half the actual duration of the analysis process was acquired and the preliminary tests carried out. Presently, noise associated to the MCA unit coupling is degrading the CdTe detector resolution, so it is not yet under use.

### Published, accepted or in press work

1. O.R. Oliveira, Relatório de Actividades, 1 de Abril 2000 a 1 de Abril 2002, ITN.
2. S.M. Almeida, M.C. Freitas, M.A. Reis, Quality Control of Air Particulate Matter PM10 and PM2.5 by  $k_0$ -INAA, *Journal of Radioanalytical and Nuclear Chemistry*. (in press)



# Gamma Radiation Treatment of Food Industry Provenance Wastewater

M.G. Ventura, M.C. Freitas, P. Matos, M.L. Botelho

## Objectives

It was carried out a study, with the purpose of determining the influence of  $\gamma$  radiation absorbed dose on the dynamic of chemical elements between solid and liquid phase present on wastewaters samples. Instrumental Neutron Activation Analysis (INAA) was used as analytical technique. For this purpose effluent from food industry (vegetables processing) was used.

## Results

Samples were irradiated in a Cobalt source (Physics Sector) with a dose rate of 0.8 kGy/h. Doses of 5 and 10 kGy were applied.

The three effluent samples of 80 ml each were filtered and two phases were obtained, a liquid and a solid phase (filtrate). The liquid phase was freeze-dried and a powder was obtained. All samples on solid phase were placed in high purity polyethylene boxes and irradiated during 30 seconds ( $\phi_{\text{term}} = 2.60 \times 10^{12}$  neutron/cm<sup>2</sup>.s. Gamma-spectra of the irradiated samples were collected using hyperpure germanium detectors of high resolution, after 8 and 30 minutes. Concentrations were determined by the  $k_0$ -standardized instrumental neutron activation analysis. Results for four elements are presented on figure 1. Apart from magnesium and manganese whose contents seem to be modified by irradiation dose, other elements like potassium and sodium remain practically constant on the two phases.

## Published, accepted or in press work

1. M.G. Ventura; *Report of Activities April 2001-April 2002*, ITN/02.
2. M.L. Botelho, M.C. Freitas, Paulo Ferreira, Sandra Cabo Verde, Márcia Ventura, Luisa Alves, Alexandre Costa, Paula Matos; *Impact of  $\epsilon$  Beam and  $\gamma$  Radiation on the Treatment of Wastewater and Drinking Water: Comparative Studies*; 1<sup>st</sup> Research Co-ordination Meeting of the IAEA - Workshop on "Wastewater Treatment by Ionizing Radiation"; October 2002, Sacavém, Portugal.

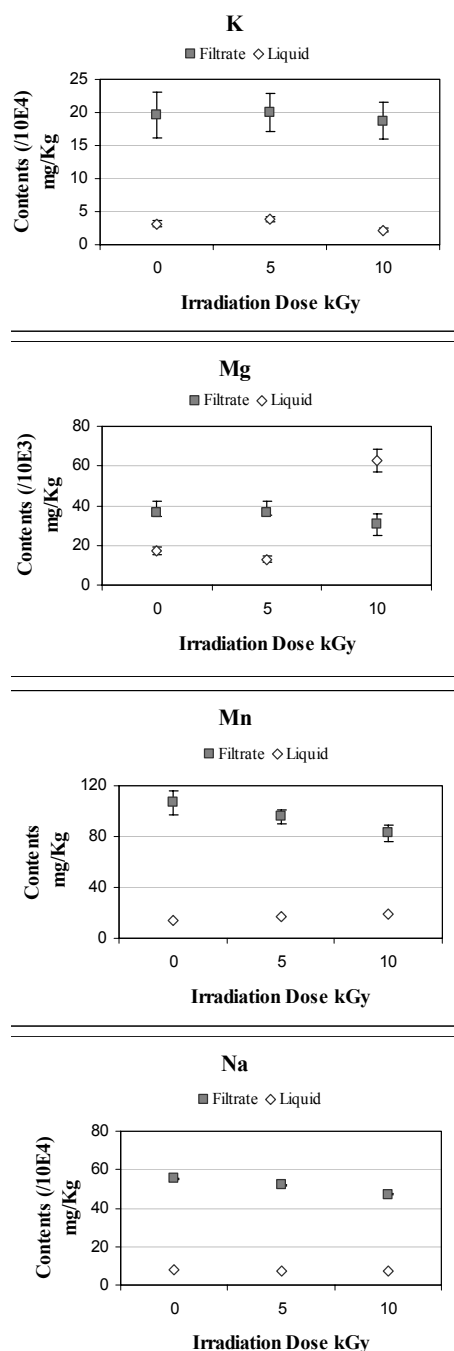


Fig. 1 K, Mg, Mn and Na contents evolution in function of irradiation dose

