# Physics Sector

*Maria Fernanda da Silva*

The Physics Sector houses three large infrastructures which are unique in Portugal:

**1-The Ion Beam Laboratory** with a 3.1 MeV Van de Graaff Accelerator and the 210 keV high fluence ion implantor. The accelerator replaced, in 1992, the old 2 MeV van de Graaff, which had been installed in 1958, in this Campus.

The strategy adopted a few years ago (in fact, since 1981) to open the infrastructure to the University and other research institutions in order to be fully used for training and education of students and young researchers, originated a strong collaboration between the Nuclear Physics Centre of the University of Lisbon (CFNUL) and ITN. With this collaboration it was possible to prepare and present a proposal for the acquisition of the **implantor** and later on of the **ion microprobe** and in this way to install the Ion Beam Laboratory as it is today.

This infrastructure is used in the fields of material sciences, environmental health, atomic and nuclear physics (cross section measurements). This last subject is carried out by a research group**, Nuclear Reactions Group**, formed with staff from the Universities of Lisbon. The activity developed in the Ion Beam Laboratory is presented in the following pages under the names **Nuclear Solid State Physics using ion beams**, **Nuclear Reactions** and **Health and Environmental studies using ion beams**.

**2- The High Temperature Materials Laboratory, Ma$^3$t** reached maturity during this second year of activity. Nevertheless, the development work at the Hotbird continued both in the software and the hardware fronts. The main development consisted in the addition of a new geometry, the *high-intensity mode* to complement the existing ones: the *double-crystal* and the *triple-axis diffractometer* geometries.
During 2001 emphasis was put on the tackling of problems that cannot be solved by conventional diffractometers. The current research projects are centred on the study of advanced materials used in the electronic industry as well as high-temperature alloys used in the aeronautical industry. Many of these studies are performed in collaboration with foreign research groups that request beam time at Hotbird owing to its high specificity and enhanced capability to solve difficult problems, e.g. the characterisation of the strain state, thickness and composition of nanometer-thick buried layers in SiGe devices.
Most of the activity developed in this Laboratory is presented under the title **Condensed Matter Physics**. Also under this name are presented the activites related with the neutron scattering work. The installation of a Two-Axis Neutron Diffractometer (DIDE) equipped with a "banana" multidetector was completed. A small Angle Neutron Scattering Instrument (SANS) is currently under installation.

**3-The Gamma Radiation facility (UTR)** is a Cobalt-60 facility with a semi-industrial dimension, which was constructed in collaboration with the International Atomic Energy Agency (IAEA), as a demonstration unit for the national industry.

For logistical reasons the unit is under the Physics Sector coordination. Its operation started in Nov.1988 with an activity of approximately 300 kCi. Today the irradiations are carried out with an activity about 56kCi.

The main activity developed around this infrastructure relates with the radioesterilisation of medical devices and pharmaceuticals and the decontamination of other products. A new field is presently being developed to apply ionising rediation techniques to waste water treatment. The research carried out is presented under the title **Radiation Technologies Processes and Products**. This activity, together with the work carried out by the **Nuclear Instruments and Methods Group**, is presented under the umbrella Industrial Technologies.

# Structure of the Sector and Technical staff

## Research groups in the Physics Sector

- Nuclear Solid State Physics using Ion Beams
- Nuclear Reactions
- Health and Environmental Studies Using Ion Beams
- Condensed Matter Physics
- Nuclear Instruments and Methods
- Radiation Technologies: Processes and Products

The composition of these groups is presented in the following pages.

## Administrative and Technical staff

- Ana Faria
- Diamantina Venâncio
- José António Cristina
- José Vieira Henriques
- Manuel Cabaça
- Maria Luisa Oliveira
- Maria Teresa Pires

# Nuclear Solid State Physics using Ion Beams

*Maria Fernanda da Silva*

The research activity of the Nuclear Solid State Physics Group is focussed mainly on the processing and characterisation of advanced materials using ion beam based techniques.

The main output of the activity is the training of young researchers and students and is visible in the publications of the group.

The group, formed with staff from ITN and the Nuclear Physics Centre of the University of Lisbon, is responsible for the operation of the **Ion Beam Laboratory (IBL)** in Sacavém, with the 3.1 MeV van de Graaff accelerator and the 210 kV High fluence Ion Implantor and for **the two hyperfine interactions laboratories** located at the Nuclear Research Centre in Lisbon and in Geneve (ISOLDE/CERN), respectively.

There are collaborations with other researchers from several Portuguese universities.
Also, considerable collaboration exists with foreign research teams, namely from the universities of Seville, Madrid, Bonn, Knoxville (USA), Budapest, amongst others. Most of these collaborations started through bilateral contracts and the participation in international projects.

In 2001, 52 papers were published in International Journals and 52 accepted for publication. These include doping of GaN with optical and electrically active ions as well as the structural characterisation of GaN/InGaN multilayer structures, **studies on insulators such as $LiNbO_3$ and $Al_2O_3$** started a few years ago, **synthesis of new compounds, like metastable alloys in Al , Ti**, and Mg and several silicides produced by implantation.
An **artificial neural network algorithm** was developed that analyses RBS data instantaneously.

It is worth mentioning the potential of the ion beam techniques for studies of thin films and multilayers. Relevant work continues to be done in the characterisation of **magnetic thin films for magnetic spin valves and tunnel junctions.**

The group participates in the **European Fusion Development** agreement technology work programme through several tasks, namely the study of compatibility of $SiC/SiC_7$ with other structural materials and surface studies of ceramic breeders ($Li_4 SiO_4$ and $Li_2 TiO_3$) and neutron multiplier (Be) after exposure to fusion relevant working conditions.

The group works also at **ISOLDE/CERN** with the $e^-$-$\gamma$ Perturbed Angular Correlations and the Emission Channeling techniques, applied to studies of High-Tc superconductors and of colossal magnetoresistive oxides with radioactive isotopes and of lattice site location of transition metals in semiconductors.

The main results of the research developed during 2001 are summarised in the following pages.

# Nuclear Solid State Physics Using Ion Beams

## Research Team

### Researchers(*)

- M.F. DA SILVA, Principal Researcher, Group leader
- E. ALVES, Auxiliary researcher
- R.C. DA SILVA, Auxiliary researcher
- L.C. ALVES, Research assistant (75%)[1]
- A. KLING, Auxiliary researcher , Reactor (5%)
- J.G. MARQUES, Auxiliary researcher, Reactor (5%)
- N. BARRADAS, Auxiliary researcher, Reactor (5%)
- J.C. SOARES, Full Professor[2]
- A.A. MELO, Associate Professor[3]
- M.R. DA SILVA Auxiliary Prof., IST
- M. BREESE, Invited Scientist (10%)
- J.G. CORREIA, ITN Contract
- A. PAÚL, PRAXIS Post-Doctoral
- L. CHANG, PRAXIS Post-Doctoral, until March 2001
- U. WAHL, PRAXIS Post-Doctoral, since Nov. 2001

### Students

- A.R. RAMOS, PhD student, PRAXIS grant until March 2001, PRAXIS post-doctoral since then.
- L. PRUDÊNCIO, PhD student, PRAXIS grant
- A. LIMA, PhD student, Univ. Aveiro, PRAXIS grant
- J.P. ARAÚJO, PhD student, Univ. Aveiro, PRAXIS grant
- C.P. MARQUES, MSc student, PRAXIS grant
- M. VILARIGUES, MSc student, PRAXIS grant
- E. RITA, Last Year BSc Student, PRAXIS grant
- V. MATIAS, Last Year BSc Student, PRAXIS grant
- A.C. MARQUES, Last Year BSc Student, PRAXIS grant

### Technical Personnel

- J. ROCHA
- F. BAPTISTA
- P. MARQUES

(*) Also members of CFNUL.
[1] Doing PhD.
[2] On leave from University of Lisbon.
[3] Retired from University of Lisbon.

## Funding

| | $\times 10^3$ PTE |
|---|---|
| Research Projects: | 46 537 |
| **Total:** | **46 537** |

## Publications

| | | | |
|---|---|---|---|
| Books: | | 1 | |
| Journals: | | 45 | and 54 in press |
| Proceedings: | | 6 | |
| Conf. Communications: | | 11 | |
| Other publications: | | 3 | |
| Theses: | PhD | 1 | |
| | MSc | 1 | |
| | BSc | 2 | |

# Thin Films and Multilayer Structures

E. Alves, M.F. da Silva, J.C. Soares, N. Franco[1], A.D. Sequeira[1], L. Rebouta[2], S. Pereira[3], R. Correia[3]

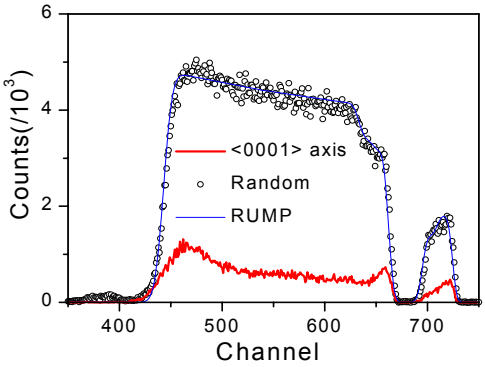
## Objectives

The ion beam laboratory of ITN runs several collaborations with other institutions in the field of thin films. The role of ITN is to provide information on parameters like composition, thickness, interface roughness, epitaxy and crystalline quality of the films.

## Results

The work carried out during this year in this area was concentrated mainly in two systems: Semiconducting films [1-11] and metallic coatings [12-13]. The research work in Semiconductors was mainly concentrated in GaN/InGaN system. [2-8,10,11]. The performance of InGaN-based devices is responsible for the intensive research developped during the last decade. The possibility to tune the wavelength emission from the UV to the green spectral regions through the control of the In content explains the technological importance of this material. Despite all these efforts some key topics like role of composition, strain and phase separation on the optical properties are still under debate.

Our purpose is to study and correlate the composition gradient and strain with the optical properties of the InGaN films. The capabilities of ion beam techniques to provide information on these issues is illustrate in the figure.




The figure shows the random and <0001> aligned spectra of an InGaN/GaN epilayer where the gradient of the In concentration is evident. The good quality of the film is also revealled by the low value of the ration between the random and aligned spectra.

The work carried out on metallic coatings aimed at study the composition and thickness of the films.

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# Doping and study of semiconductors with ion beams

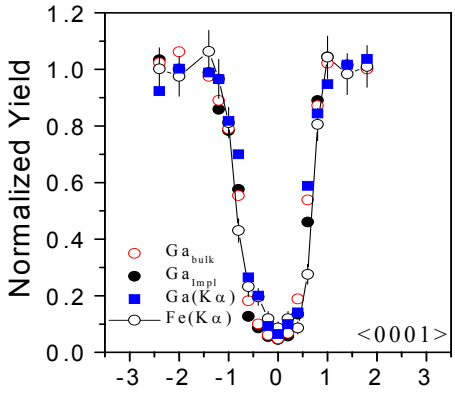
E. Alves, C. Liu, M. da Silva, J.C. Soares, N. Franco[1], A.D. Sequeira[1], K. Lorenz[2], R. Vianden[2], T. Monteiro[3], M.J. Soares[3]

## Objectives

The main goal of this work was doping GaN with optical active ions. The role of ITN in this collaboration is to provide information on damage recovery and lattice site location of the dopants.

## Results

We implanted rare earth (RE) [1-7] and metallic [8-1] ions in GaN. Doping GaN with RE extends its operating wavelenght to the infrared, green and red region of the optical spectra. However, integrating ion implantation with the GaN technology depends on being able to anneal the implantation damage. We studied the influence of implantation parameters (temperature and dose) on the damage production and determined the best annealing conditions. We also followed the lattice site location of the implanted ions. An example of the capability of ion beam techniques is shown in the figure, where the angular scan along the <0001> axis shows the complete overlap of Fe and Ga x-ray curves indicating that Fe is substitutional along this direction. A similar result found along the <10-11> axis clearly shows that Fe occupies mainly the Ga site in the lattice [7].




***X-ray angular scan along the <0001> axis revealing***

## Published, accepted or in press work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# Synthesis of New Compounds by Ion Implantation

R.C. da Silva, A.R. Ramos, L.M. Prudêncio, M. Vilarigues, N.P. Barradas[1], J. Salvador[2], O. Conde[3], R. Vilar[2], F. Pászti[4], É. Vázsonyi[4], A.D. Sequeira[5], M.R. da Silva[2], M.F. da Silva, J.C. Soares

## Objectives

Ion beams can be used to modify the surface of technologically interesting materials, without altering its bulk properties. In some cases novell phases appear. Our work concentrates on the ion beam synthesis in semi-conductors (cristallyne and porous silicon) and metals (Al-Cr, Al-Fe, Ti-Cr, Ti-Al, Mg-Cr).

## Results

***1. Ion beam synthesis of silicides:***

***1.1. In porous silicon***

High dose implantation was used to form polycrystalline silicide films on porous silicon (PS) with different native concentrations of C and O impurities. Results show that the light impurities were partially expelled from the forming silicide layer. Combining XTEM with ion beam methods it was possible to show that, in the implanted region, the porous structure collapses and densifies during implantation, but the underlying porous silicon remains intact. The layer structure, as well as the quality and type of the formed silicide, were found to depend on the original impurity level, implantation temperature and annealing [1,2].

***1.2. In crystalline silicon: the influence of the O content in the synthesis of $HfSi_2$***

Float zone (FZ) and Czochralski (CZ) Si wafers were simultaneously implanted with $Hf^+$ and subsequently annealed. We showed that the resistivity changes differently with annealing in the FZ and CZ samples, though the final values do not depend on the oxygen content. Results obtained in O and Hf co-implanted samples show that, despite the fact that Hf is an O getterer, the $HfSi_2$ film still forms, expelling the O, which accumulates at the silicide /substrate interface.

***2. Ion beam synthesis in metals:***

***2.1. In Aluminium***

***2.1.1. Ion Nitriding***

We have studied the kinetics of Al ion nitriding under different substrate temperatures. Depending on the experimental conditions, the nitriding kinetics are either controlled by the delivery of N ions or by the diffusion of Al atoms. The growth of the nitride layer is limited by poor layer adhesion. An hexagonal AlN-phase plus a small fraction of the cubic AlN-phase are formed [3].

***2.1.2. Cr and Fe implantation***

High purity polycrystalline Al discs were implanted either with Cr or Fe, at different substrate temperatures.

***Al-Cr system***: three different types of surface layers form after Cr ion implantation in Al. At RT a solid solution forms. By implanting around 270ºC and 500ºC, the intermetallic $Al_{86}Cr_{14}$ and $Al_{13}Cr_2$ form respectively. Corrosion resistance tests show that the Al-Cr solid solution is the noblest system, followed by $Al_{86}Cr_{14}$ and $Al_{13}Cr_2$. All these systems are nobler than pure Al.

***Al-Fe system***: RBS and CEMS show that the $Al_5Fe_2$ intermetallic forms during implantation. After 1 hour annealing at 630ºC it transforms to $Al_{13}Fe_4$ [4].

***2.2. In Titanium***

High purity polycrystalline Ti discs were implanted either with Cr or Al, at different temperatures.

***Ti-Cr system***: no Ti-Cr intermetallic compound forms, either after ion implantation or after annealings up to 1000 ºC. Very fast Cr transport was found to occur at temperatures above 600 ºC, precluding the formation of intermetallic compunds.

***Ti-Al system***: no Ti-Al intermetallic compound has been formed so far.

***2.3. In Magnesium***

High purity polycrystalline magnesium was implanted with $Cr^+$ ions to promote corrosion resistance. The results obtained so far show that: *a*) corrosion takes place at a slower rate in the Cr implanted Mg surfaces; *b*) there is a dose effect and a dose for which the effect is maximum; *c*) $Mg(OH)_2$ forms, along with hydrated species of $MgCrO_4$ and chlorine containing compounds. Although the detailed mechanism is not known at present, the formation of Cr containing Mg (mixed) oxide must be responsible for slowing down the progress of corrosion attack by the Cl ions [5].

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4. L. Prudencio, J.C. Waerenborgh, A.P. Goncalves, O. Conde, R.C. da Silva, Formation of Al-Fe surface alloys by ion implantation of Fe in Al, *Surf. and Coat. Technol*, *in press.*
5. M. Vilarigues, L.C. Alves, N. Franco, A.D. Sequeira, R.C. da Silva, Characterization of corrosion products in Cr implanted Mg surfaces, *Surf. and Coat. Technol*, *in press.*

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[3] FCUL - Faculdade de Ciências da Universidade de Lisboa, Dep. Física.
[4] KFKI-Res. Inst. for Particle and Nuclear Physics, Budapest, Hungary and MTA- Res. Inst. for Technical Phys. and Materials Science, Budapest, Hungary.
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# Experimental Development

L.M. Redondo[1,2], E. Margato[1,3], J. Fernando Silva[3],
J. Rocha[4], N. Pinhão[4], J.C.Soares

## Abstract

A modular concept on high-voltage pulse generators is under development for future use in facilities for plasma immersion ion implantation (PIII). The generator proposed uses individual modules each one consisting of a pulse circuit based on a step-up transformer; the secondary of each step-up transformer is connected in series. Each step-up transformer delivers a fraction of the total voltage with primary voltage supplied via an isolation transformer. With this topology we expect to achieve tens of kV with low voltage semiconductor switches (<1kV). A three 5kV-module initial prototype was assembled with 800V semiconductor switches and experimentally tested for an output of 11kV, 5μs pulse width and 10kHz-pulse frequency. Different load conditions were already tested.

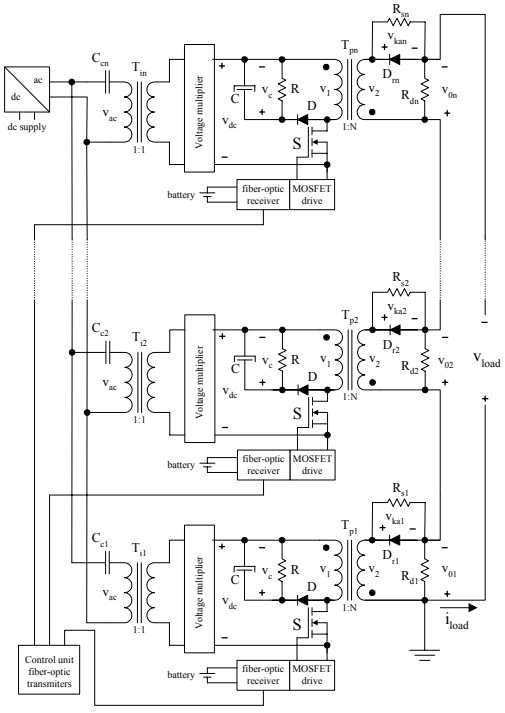


**Figure 1.** Modular concept high-voltage generator circuit.

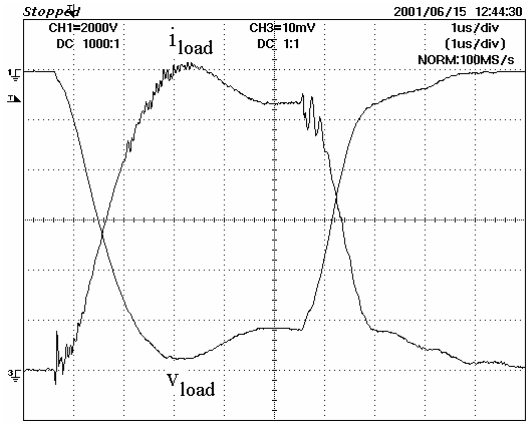


**Figure 2.** Measured waveforms of the pulse with approximately 11kV and 1.2A current, obtained at the terminals of sries connected carbon filament lamps. Time basis is 1μs/div, $v_{load}$ (2kV/div), $i_{load}$ (0.2A/div).

## Published, accepted or in press work

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4. L.M. Redondo, E. Margato, J. Fernando Silva, Rise time reduction in high-voltage pulse transformers using auxiliary windings, *Transactions on Power Electronics*, in press.

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# Nuclear Microprobe Studies

L.C.Alves, M.B.H. Breese[1], E. Alves, A. Paúl, E.M.C. Rita, M.A. Reis, J. Ferreira[2], M. O. Figueiredo[3], M.R. da Silva, M.F. da Silva, J.C.Soares, F. Rodrigues[4], P. Fialho[4]

## Objectives

To continue and improve analytical methodologies in the fields of material science and biomedicine. Extend the microprobe work to the characterisation of air particulate matter and geological samples.

## Results

Among the studies performed in material science, a significant amount of time was devoted to the characterisation of new materials for fusion reactors [1]. Included were the new $SiC/SiC_f$ 4D composite materials produced by CERASEP® (elemental impurity distribution and contents), Be pebbles (elemental characterisation and surface oxygen distribution) and lithium orthosilicate compounds.

In collaboration with the University of Azores, individual particles were analysed in aerosol samples with two different provenances (Sahara Desert and North America) and some distinguishable features were established. Based on the relative elemental abundance of the elements, obtained by combining PIXE and RBS analytical techniques, the existence of certain compounds could be proposed (NaCl, $CaCO_3$, $CaSO_4$ $Fe(OH)_3$ and a molecule with Cu:Cl in a 1:2 ratio) [2].

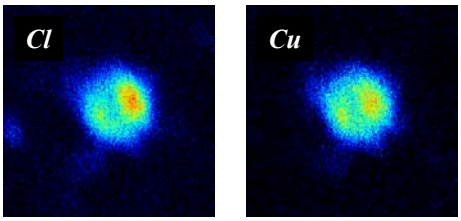


**Fig. 1** – X-ray elemental maps from a 50*50μm$^2$ scan of an aerosol sample.

As a new research field in the Ion Beam Laboratory, and with the IGM collaboration, exploratory work is being done in the analysis and characterisation of pyrite and arsenopyrite samples. In fig. 2 are shown the elemental maps obtained from an arsenopyrite embedded in a quartz matrix. Chemical reactions with hydrothermal fluids can be observed resulting in the solubilization of S and the transport and precipitation of Fe and As.

Technical implementation was also undertaken with emphasis in a Secondary Electron Image System for improving topographic image contrast. The system distinguishes 1 μm high features, improving the sample positioning capabilities and the identification of particular features [3].

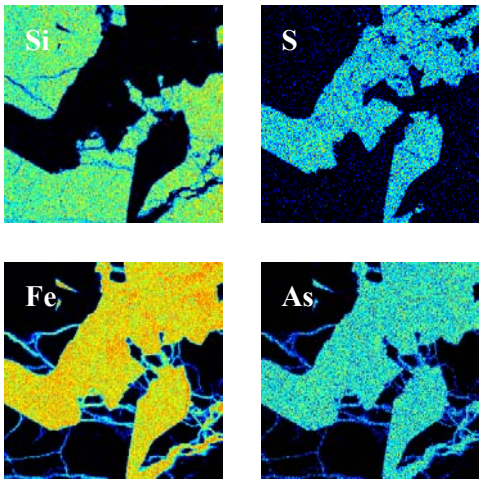


**Fig. 2** – X-ray elemental maps from a 2640*2640μm$^2$ scan of an arsenopyrite sample.

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# Advanced Data Analysis of Ion Beam Analysis Data

N.P. Barradas, A. Vieira[1], C. Jeynes[2]

## Objectives

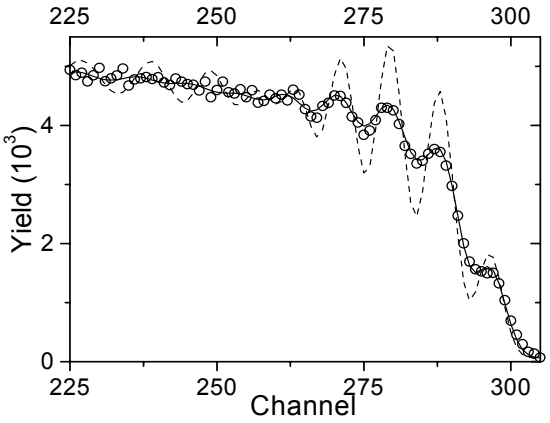
Ion Beam Analysis (IBA) is a cluster of techniques dedicated to the analysis of materials. Data analysis is normally done in an interactive way, requiring a dedicated expert. Our goal is to automate IBA data analysis.

## Results

We have previously developed a code, the IBA DataFurnace, that uses the simulated annealing algorithm to automatically analyse IBA data. We have now extended that code to be able to analyse data of samples where interfacial roughness is present. This is a complex problem, and it is the first time that generation of spectra including rougness can be done in a simple and fast way [1-4].

We have also opened a new line of work, namely using artificial neural networks (ANNs). We have developed ANNs both to analyse data as such, extracting the relevant desired parameters in an instantaneous way [5-8], and in a code suitable for automated control of an experimental setup, given an interface to the relevant hardware [9].

The determination of the stopping power of ions in matter is experimentally intensive and difficult. We developed a method to determine stopping powers from simple Rutherford backscattering spectra, using Bayesian inference, and applied it to the important system $^4$He in Si. The results compare very well with other measurements. A 1% accuracy is achieved [10].




Fit (solid line) and data (circles) obtained for a sample MgO/(Fe23Å/Co20Å)$_{10}$. A simulation made assuming no interfacial roughness (dashed line) is also shown.

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# Magnetic Spin Valves and Tunnel Junctions

N.P. Barradas, S. Cardoso[1], P.P. Freitas[1], U. Kreissig[2], A.R. Ramos, M.F. da Silva, J.C. Soares, J. Wang[1], P. Wei, Z. Zhang[1]

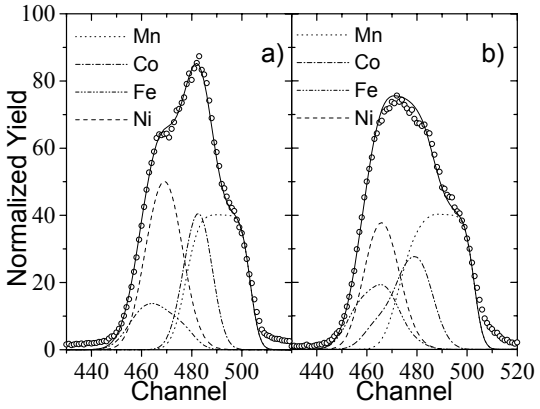


***Fig.*** Detailed He-RBS spectra (open circles) and simulations (solid lines) of the Mn, Fe, Ni and Co signal for the a) as grown and b) annealed sample.

## Objectives

ITN has a long-standing collaboration with the INESC magnetic systems group led by Prof. Paulo de Freitas. The role of ITN is to provide structural characterisation of the highly complex advanced magnetic systems produced at INESC. This is a highly interactive collaboration that has proved to be very successful.

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3. S. Cardoso, Z. G. Zhang, P. P. Freitas, P. Wei, N. Barradas, J. C. Soares. Electrode roughness and interfacial mixing effects on tunnel junction thermal stability, *J. Appl. Phys.* 89 (2001) 6650-6652.
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## Results

This year the work was concentrated in improving the characteristic of spin tunnel junctions [1-9]. These are made by two thin magnetic layers separated by a thin insulating one. The tunnelling current across this barrier layer depends on the relative alignment of the magnetisations of the two magnetic layers. Hence such a device can be used either as a reading head (in which the external field - e.g. from a hard disk - leads to a change in the resistance of the system), or as a non-volatile memory (where the external field forces the direction of the magnetisation).

The highlights were to increase the annealing temperature that the tunnel junctions can withstand before losing their quality, which is essential for integration with standard device technology, and to lower the resistance of the insulating junction, which leads to a higher magnetoresistance signal.

To illustrate the capacities of ITN in providing fundamental information, the figure shows the metal part of the RBS spectrum of a $Si/Al_2O_3$ 750Å/NiFe/CoFe/$Al_2O_3$ 15Å/CoFe/MnIr 250Å/Ta30Å sample, as grown and annealed at 435ºC. The changes can be assigned to diffusion between certain interfaces. The junction design has been altered taking this information into account.

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# $e^-$-γ Perturbed Angular Correlations and Emission Channeling at ISOLDE/CERN

E. Alves, V. Amaral[1], J.P. Araújo[2], J.G. Correia, Y. Li[1], A.M. Lopes[1], A.C. Marques, J.G. Marques[3], A.A. Melo, A.R. Ramos, M. Reis Jr.[1], E. Rita, V.M. Serrão, M.F. da Silva, J.C. Soares, U. Wahl and the ISOLDE collaboration[4]

## Objectives

The Nuclear Solid State Physics Lisbon group (ITN, CFNUL) works at ISOLDE since 14 years. There we have introduced and developed the $e^-$-γ Perturbed Angular Correlations ($e^-$-γ PAC) and, more recently, the Emission Channeling Techniques (EC), working with new 2D electron detectors. PAC and EC are powerful tools on characterizing the microscopic behavior of elements complementarily to the ion beam techniques at ITN-Sacavém. From such developing work new PAC probes are recently available [1,2].
The scientific work is actually centered in three research subjects approved by the ISOLDE Scientific Committee (INTC) from 11 / 2000 to 02 / 2001.

## Results

*a) Studies of Colossal Magnetoresistance Oxides with Radioactive Isotopes (Project IS390)*
Studies on $La_{1-x}Cd_x$ $MnO_3$ pellets and thin films, show very different (and unexpected) macroscopic magneto-electric properties from $La_{1-x}Ca_x$ $MnO_3$. Preliminary PAC results, obtained for $^{111m}$Cd-implanted $La_{1-x}Cd_x$ $MnO_3$, show the presence of a magnetic field of about 3T at the Cd site, below the Curie temperature, around 180K [3]. These are surprising results, since strong magnetic fields are only expected to occur at the Mn sites. Fits of the emission channeling patterns to the experimental data showed that Cd preferentially occupies the Mn site for doping concentrations x > 0.25. Further PAC results performed with $^{111}$Cd on $LaMnO_{3+\delta}$ did detect rhombohedral to orthorhombic and orthorhombic-prime phase transitions with great accuracy, as a function of the oxygen concentration.
*b) Studies of High –Tc Superconductors Doped with Radioactive Isotopes (Project IS360).*
Due to the ISOLDE breackdown during 2001 a complementary study has been presented onto the carbon contamination and stoichiometry of pellets and thin films of Hg- $Hg_xTl_{1-x}$ and Tl- based high-Tc superconductors. [4].

*c) Lattice Site Location of Transition Metals in Semiconductors (Project IS368).*
We have shown that Fe is substitutional on GaN at the Ga site, proving that the absence of deep acceptor centers cannot be due to Fe on non-substitutional sites [5]. On Si, the implanted Ag occupies near-substitutional lattice sites [6], further indicating that the formation of near-substitutional Ag is, at least partly, responsible for the defect gettering effect of Ag in Si. Further experiments have shown that Cu, Er, In and Lu are subsititutional on the wide band gap ZnO semiconductor.
*d)* A contract has been established with a high-tech industrial company, ITN (Sacavém), IKS (Leuven) and ISOLDE/CERN to identify the lattice site location of Cu in KLiTaNb, a new material with optoelectronic applications. Only EC can provide such information. The main difficulty concerns the simulation of the EC patterns, which we have recently proven to be possible, with experiments of lattice site location of Er in $SrTiO_3$, with a similar structure to KLTN [7].

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2. W. Tröger, *et al*., "The Nuclear Quadrupole Interaction of 204mPb in Cadmium monitored by γ–γ Perturbed Angular Correlations*", Zeitschrift für Naturforschung*, in press.
3. J.P. Araújo, *et al*., "Hyperfine Fields at the Cd Site in La0.67Cd0.25MnO3 CMR Manganites", *Hyperfine Interactions* 133 (2001) 89.
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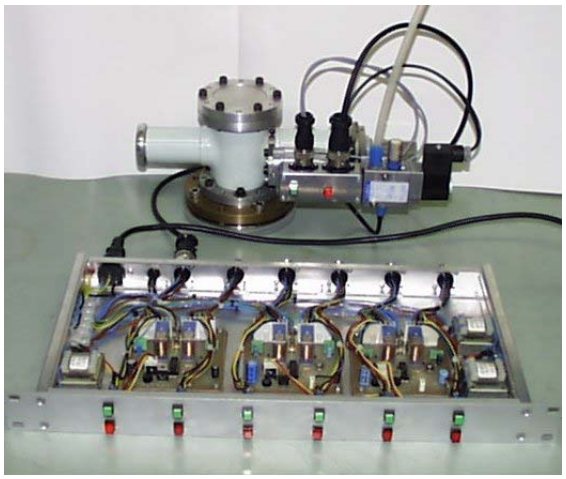
[2] Physics Institute Univ. Porto, Portugal
[3] Reactor, IT, Sacavém, Portugal
[4] ISOLDE, CERN, Geneva, Switzerland.

# Laboratory Operation and Development

M. Ribeiro da Silva

**3 MeV Van de Graaff Accelerator**
**Beam Lines**

A remote_local controller for the different beam stoppers has been installed in the Van de Graaff and experimental lines. The beam stoppers are compressed air powered and locally or remote controlled through a central console mounted in the accelerator control rack. Indicator lights, both local and remote, give a visual indication of the stoppers position.



Beam stopper and central control unit

**New Laboratory Equipment**

In the scope of the "Breeding Blanket Development (European Fusion Technology Programme)" a new experimental set-up was developed and installed. It consists in a resistivity measurement unit that allows measurements of the resistivity of non-massive samples (powder or pebbles) in a range of temperatures up to 850 ºC and variable pressure (up to 5000 n/cm$^2$). The measurements can be done in a controlled gas flow. In figure 1 we can find the layout of the set-up.

**General purpose Power Supply**

For the resistivity measurement unit and as a general laboratory unit a new controlled power supply was developed and constructed. This unit presents the following parameters:

- max. output power 1,6 kW, max. current 8A;
- stabilized alternate voltage output from 0 to 240 V; the output power is stabilized by a microprocessor PID controller with coupled thermocouple (K, J, N or platinum);
- neutral fase detection and blocking;
- power supply outlets adapted to the different plug standards;
- full visualization of the functioning status is available;

In figure 2 we can find a general view of the instrument.

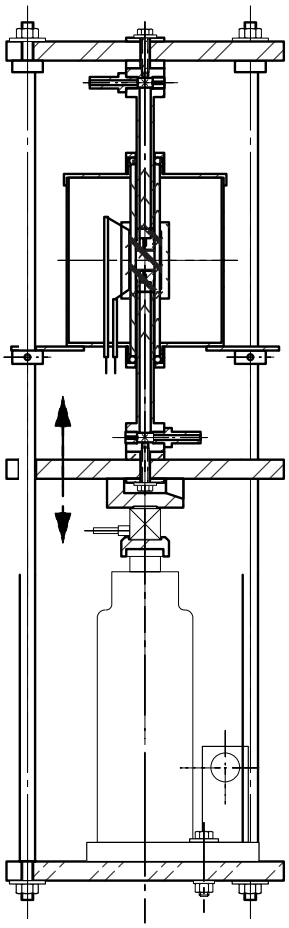


Fig. 1 - Resistivity mesurement unit

Fig. 2 - General pourpose Power Suply

# University Group of Nuclear Reactions

*Adelaide Pedro de Jesus*

This group has been involved in the study of proton-induced nuclear reactions with the objectives to obtain cross sections of nuclear reactions relevant to nuclear astrophysics and to extend analytical capabilities to light elements.

So far, the experimental work has relied upon ITN – Ion Beam Laboratory, based on a 2.5MV Van de Graaff accelerator. This facility has allowed the development of an accurate method to measure in an absolute way cross sections of relevant nuclear reactions. Also for the applied point of view an effort has been done to complement the already installed PIXE facility by developing a set-up of PIGE analysis for light elements.

In order to proceed with experimental work related to astrophysically relevant nuclear reactions it is imperative to be able to measure the respective cross sections at very low energies, where the cross sections are lower than the picobarn. At these energies several challenges must be overcome: the effects of cosmic rays in the detectors must be reduced to almost zero with an adequate detector shielding, the Coulomb barrier is partially screened by the atomic electron cloud surrounding the nuclei, which leads to an enhanced cross section compared with bare nuclei cross section; stopping powers are not well known. So, we have joined LUNA (Laboratory for Underground Nuclear Astrophysics) collaboration. Work under LUNA collaboration has been centred on the study of electron shielding effects on nuclear reaction cross sections at very low energies and on the study of the reactions $d(p,\gamma)^3He$ and $^{14}N(p,\gamma)^{15}O$. Target preparation and stoicheiometry analysis by ERD, RBS, PIGE and NRA, has been done at ITN.

In the short term the work to develop a calibrated PIGE set-up will be concluded, opening new perspectives in apllied work for Environment, Materials and Health Sciences and Geology. Also, work under LUNA collaboration will proceed with the study of relevant reactions of the p-p chain and CNO cycles. This collaboration will imply some experimental work at ITN, namely for target preparation and stoicheiometry analysis by ion beam techniques and cross section measurements for normalisation of LUNA measure-ments.

In the medium term, the acquisition of a new accelerator would open new research areas within nuclear astrophysics, as the study of relevant reactions of the p-process nucleosynthesis.

# University Group of Nuclear Reactions

## Research Team

### Researchers

- ADELAIDE P. DE JESUS, Full Professor (FCT/UNL), Group Leader
- J. PIRES RIBEIRO, Associate Professor (FCUL)

### Students

- BRUNO BRAIZINHA, PhD Student, FCT grant
- JOÃO CRUZ, PhD Student, (FCT/UNL)
- RODRIGO MATEUS, PhD Student, FCT grant
- JOANA VAZ PINTO, BIC PRAXIS XXI

## Funding

| | $\times 10^3$ PTE |
|---|---|
| Research Projects: | 9 100 |
| **Total:** | **9 100** |

## Publications

| | |
|---|---|
| Journals: | 1 and 1 in press |
| Proceedings: | 1 |
| Conf. Communications: | 1 |

# Experimental Study of Nuclear Reactions for Astrophysics

A. P. Jesus[1,2] J. Cruz[1,2] B. Braizinha[2] J.V. Pinto[1,2] J. P. Ribeiro[2,3]

## Objectives

Our understanding of the evolution of the very early universe depends crucially on modelling the primordial nucleosynthesis of the light elements. Calculations of primordial and stellar nucleosynthesis are strongly affected by uncertainties in the nuclear input data. In order to lower the uncertainties of the astrophysical factor of relevant reactions of the p-p chain and CNO cycles, a research program was established involving:

1. Development at ITN of an experimental method to determine in an absolute way the reaction cross sections.
2. Experimental work on reaction cross-sections at relevant energies (around the solar Gamow peak) under LUNA (Laboratory for Underground Nuclear Astrophysics) collaboration.

## Results

1. The experimental method to determine in an absolute way the reaction cross sections was applied to the study of proton induced reactions on fluorine, namely, $^{19}F(p,p'\gamma)^{19}F$, $^{19}F(p,p)^{19}F$ [1], $^{19}F(p,\alpha\gamma)^{16}O$ [2], $^{19}F(p,\alpha)^{16}O$ [2], and to the study of reactions on $^{7}Li$, namely, $^{7}Li(p,p)^{7}Li$ , $^{7}Li(p,p'\gamma)^{7}Li$ [3], $^{7}Li(p,\gamma)^{8}Be$, $^{7}Li(p,\alpha)^{4}He$ (fig.1). These two last reactions are being study theoretically and results both experimental and theoretical will be published together soon.
2. Work under LUNA collaboration has been centred on the study of electron shielding effects on nuclear reaction cross sections at very low energies and on the study of the reactions $d(p,\gamma)^{3}He$ and $^{14}N(p,\gamma)^{15}O$. Target preparation and steocheiometry analysis by ERD, RBS, PIGE and NRA, has been done at ITN.

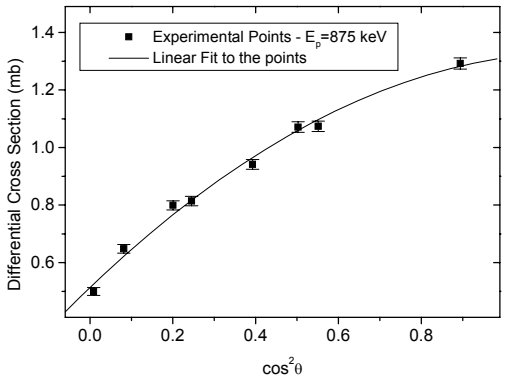



Fig. 1 Angular distribution of alpha-particles from $^{7}Li(p,\alpha)^{4}He$, reaction, obtained for 875 keV protons.

## Published, accepted or in press work

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3. R. Mateus, A.P. Jesus, B. Braizinha, J. Cruz, J.V. Pinto, J. P. Ribeiro, "Analysis of Lithium in thick samples", accepted for publication in Nucl. Instr. Meth. B.

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# Nuclear Reactions for Applications

R. Mateus[3], J.V. Pinto[1,2], J. Cruz[1,2], J.P. Ribeiro[2,3], A.P. Jesus[1,2], A.R. Ramos[4], A.Paúl[4]

## Objectives

*Proton-induced nuclear reactions in light isotopes can be used to complement other ion beam analytical techniques. Proton-Induced Gamma-ray Emission, PIGE, allows the detection and quantification of F, Li, Be, B, Na, Mg, Al, extending the analytical capabilities of PIXE. Elastic scattering cross sections may be several orders of magnitude larger than Rutherford cross sections, as is the case for C, N, O, Al, Si, leading to a better sensitivity than alpha-RBS. Together with a very good energy and depth resolution, this makes proton elastic scattering a powerful tool for light element analysis.*

PIGE analysis of thick and intermediate samples is usually performed with the help of standards, but this method gives only good results when the standard is very similar to the sample to be analysed. A more precise method can be implemented based on a code that integrates the nuclear reaction excitation function along the depth of the sample. For that purpose, it is necessary to measure the reaction excitation function in the exact analytical conditions. The energy steps needed to define accurately the excitation function can be used as energy intervals for the integration procedure.

The cross sections of elastic scattering of protons by light elements are dominated by resonances and are highly anisotropic. In order to be able to quantify accurately these elements, it is necessary to measure the cross sections for all different geometric set-ups.

## Results

A code for PIGE was developed to calculate concentrations of elements in thick and intermediate samples using the element (isotope) nuclear reaction excitation function as input. This code was tested for Li and F. Excitation functions for $^{7}Li(p,p'\gamma)^{7}Li$ (Fig.1), $^{19}F(p,p'\gamma)^{19}F$, $^{10}B(p,\alpha\gamma)^{7}Be$ were obtained and introduced as input.

Thick target gamma yields for several samples containing F, Li and B were calculated and compared with yields obtained experimentally. In order to check the reliability and accuracy of the method over a very large range of concentrations, the calculated thick target yield of samples with several amounts of Fluorine and Lithium was compared with the respective measured yields. Results for Fluorine and Lithium [1] show that this method is a reliable and accurate method for PIGE analysis of F and Li in thick samples. Data analysis for B is proceeding.

The (p,p) elastic scattering cross sections of natural samples of carbon, nitrogen, oxygen, aluminium and silicon have been measured at energies ranging between 500 and 2500 keV and at laboratory scattering angles of 178° and 140°. Results are compared with previous literature data and simulations and are presented in graphical form. The measured cross-sections have been used to simulate spectra taken from known samples and have been found appropriate for quantitative calculations [2,3].

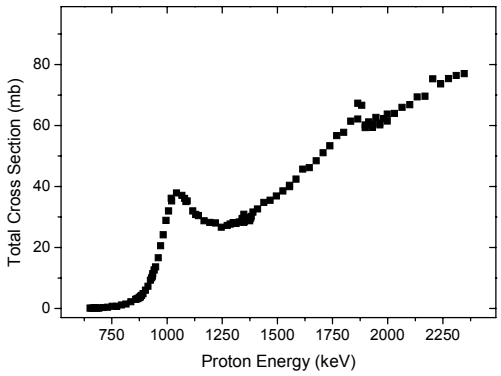


Fig.1 – Excitation function for inelastic scattering of protons by $^{7}Li$.

## Published, accepted or in press work

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2. A.R. Ramos, A. Paúl, L.Rijniers, M.F. da Silva, J.C. Soares, "Measurement of (p,p) elastic differential cross-sections for carbon, nitrogen, oxygen, aluminium and silicon in the 500-2500 keV range at 178° and 140° laboratory scattering angles", accepted for publication in Nucl. Instr. Meth. B.
3. A. Paúl, A.R. Ramos, L.Rijniers, M.F. da Silva, J.C. Soares, "Measurement of (p,p) elastic cross-sections for $^{14}N$ and $^{27}Al$ at laboratory angles of 178° and 140° in the range 0.5-2.5 MeV", *Materials Science Forum*, **383** (2002) 191

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[2] Centro de Física Nuclear da Universidade de Lisboa
[3] Department Faculdade de Ciências da Universidade de Lisboa
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# Health and Environmental Studies Using Ion Beams

*Teresa Pinheiro*

The activities at the Ion Beam Laboratory (IBL) dealing with the Health and Environmental Studies are devoted to research in ecotoxicology and molecular medicine with an emphasis on studies of the relationship of animals with the natural resource and on studies of biochemical markers in human pathological conditions.

So far, the profficiency offered by IBL to those areas is bound to the elemental and morphological characterisation of various biological matrices, that recently has been improved with the instalation of the Nuclear Microprobe at the Van de Graaff accelerator.

However, to give a confident contribution to research in ecotoxicology and in biomedicine many other aspects should be focused.

The collaborative work that is being carried out with the Centro de Biologia Ambiental and Centro de Estudos de Bioquímica e Fisiologia of the Faculty of Sciences and with the Centro de Estudos de Doenças Pulmonares of the Faculty of Medicine, all from the Lisbon University, allowed complementing common research purposes.

External funding from national and international organisations has been supporting the research activities. Projects within eco-toxicology and public health areas are running which deal with the influence of environmental pollutants or pathology conditions on redox balance and on specific biochemical markers in tissues and organs of concern.

International colaboration granted the group with new partnerships, through proposals within the European Commission RTD actions and bilateral cooperation with Spanish universities (Seville and Badajoz), opening new fields of research, specially in Biomedicine, e.g., osteoporosis and skin inflammation processes.

The co-operative work with national and international research institutions also permitted to participate in programmes for the graduation and post-graduation of students in different scientific areas, such as, Biology, Biochemistry and Applyed Physics.

The major difficulty in carrying through all the running projects and ensuring the continuation of established collaborations is the scarcity of personnel at ITN fully commited to this scientific activity. Graduation students accomplish partially the activities foreseen in the project work programmes. Although, due to the temporary character of their participation the distribution of tasks that would be advantageous for the expansion of these activities at IBL, is obviously restricted.

Also, a short-term programme for the modernisation and improvement of existent infrastructures and the investement in new analytical methodologies more versatile for research in Environment related areas should be reinforced, as referred in previous reports and repeatedly pointed out by external evaluation boards.

This measure, together with adequate human resources planning would consolidate proficiencies achieved and allow a more efficient progress of the research activities.

# Health and Environmental Studies Using Ion Beams

## Research Team

### Researchers

- T. PINHEIRO, Auxiliary Researcher (100%)
- L.C. ALVES, Research Assistant (25%)

### Collaborators

- A.M. VIEGAS-CRESPO, Associated Researcher, CBA/FFC-UL
- C. SANTOS, Associated Researcher, CEBF/FFC-UL
- A .B. ALMEIDA, Associated Researcher, Centro de Estudos de Doenças Pulmonares (CEDP)/ADIFM-UL

### Students

- C. MONTEIRO, PhD Student, ISCS/CEBF FFC-UL
- M.D.Y.ALCALÁ, PhD Student, CNA, CICA/Univ. Seville, Spain
- N. RAIMUNDO, Graduated Student, ISCS/CEBF FFC-UL
- P. NAPOLEÃO

### Technicians

- R. PINHEIRO, laboratory technician

### Funding

| | $\times 10^3$ PTE |
|---|---|
| Research Projects: | 850 |
| Other Sources: | 300 |
| **Total:** | **1150** |

### Publications

| | |
|---|---|
| Journals: | 4 and 2 in press |
| Proceedings: | 3 |
| Conf. Communications: | 1 |

# Application of Ion Beam Techniques to Biomedical and Eco-toxicological Studies

C. Monteiro[1,2], N. Raimundo[1,2], M.C. Santos[1], M. L. Mathias[3], A.M. Viegas-Crespo[3], P. Napoleão[4], C. Sousa Reis[4], A.Bugalho de Almeida[5], M.J. Palhano[5], M.D. Ynsa[6], F.J. Ager[6], M.F. Araújo[7], A.Barreiros[8], L.C. Alves and T. Pinheiro

## Objectives

The running projects and proposals for the next triennium center the research activities in the evaluation of the importance of endogeneous factors playing a role in human diseases susceptibility or expressing the response of organisms to environmental conditions. The relationship between functional and physiological changes with biochemical markers of oxidative injury and of tissue damage may contribute to the interpretation of the mechanisms involved in disease or adaptation.

## Results

### 1. Biomedical Applications

*1.1. Fate of Respired Particles in Human Respiratory System*

Evaluation of the deposition of respired inorganic materials and their removal from lungs are being studied using microscopies based on charged particles (NMP) and synchrotron radiation (SRXRF).

The figure 1 exemplifies the type and constitution of inclusions that can be visualised at alveoli walls and some aspects of elemental inclusions in phagocytic cells of the Mucosa Associated Lymphoid Tissue (MALT) [1].

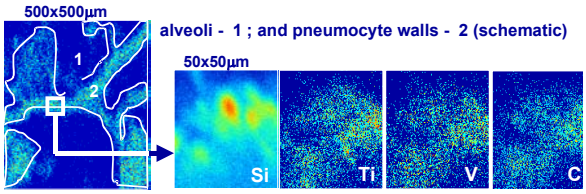


**Fig. 1** a) NMP- elemental distribution of alveoli walls

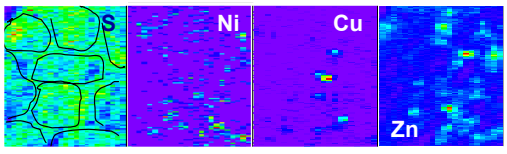


**Fig. 1** b) SRXRF - elemental distribution in MALT phagocytic cells (boundaries marked in S map); sample area scanned of 250x250µm.

1.2. *Study of the effect of steroid therapies in osteoporosis* (Cooperation with CNA-University of Seville and Fac. Medicine, Badajoz, Spain).

An animal model is used to assess bone alterations following steroid-based therapy. The study includes a PhD programme of work in Applied Phisics [2].

1.3. *Skin morphology and inflammation* (in cooperation with the Dermatology Clinic, FM-UL and the Inorganic and Radiopharmaceutical Chemistry Group-ITN).

Skin alterations due to a disease of the iron metabolism (haemochromatosis) are being studied. Nuclear microscopies, cytochemical methods and biochemical markers contribute to the study of morphological changes and of extracellular deposits of iron at epidermal regions (Fig.2).

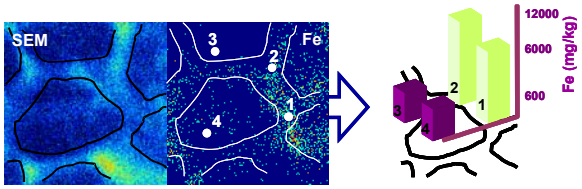


**Fig. 2**- Keratinocyte at basal epidermis, SEM image anf Fe distribution. Fe concentrations plotted: extra-cell (1,2) and intra-cell (3,4).

The team participated in a consortium for the preparation of a EC-RTD proposal (acronym, NANODERM). Modifications recommended by the evaluation board are carrying through to project re-submission at next call (February 2002).

### 2. Eco-Toxicological and physiological studies using animal models

*2.1. Biological monitoring in terrestrial ecosystems.*

JNICT/DGA/PEAM/P/NMA/666/95 project closed May 2001. Data is being evaluated and publications prepared [3,4]. The continuation of bioavailability studies progress in what concerns wild populations of small mammals to broaden the data basis to other metal pollutants.

2.2. *Biological monitoring of metal pollutants in marine ecosystems* (Biology Graduation Thesis within the work programme of the project PRAXIS XXI/2/2.1/MAR/ 170/95- 1999-2002).

The Octopus as a bio-indicator of the marine ecosystem and the study of its life cycle are investigated. Biological data are being related with elemental distributions for edible animal tissues and for vestigial shell to infer living habits and ageing, and to assess stocks for a correct use of the resource.

2.3. *Study of the Se biochemistry and the potential toxicity of selenite in rat tissues* (PhD thesis in Biochemistry ).

The effect of early Se supplementation at moderate levels is studied in rat liver, testes and blood to assess eventual interference of Se upon other elements homeostasis and upon enzyme activities related to the glutathione metabolism.

## Published, accepted or in press work

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2. Ynsa M.D., Pinheiro T., Ager F.J., Alves L.C., Millán J.C., Gómez-Zubelbia M.A., Respaldiza M.A., PIXE Studies of Osteoporosis Preventive Treatments, *Nucl. Instr. and Meth.(in press).*
3. Lopes P.A, Pinheiro T., Santos M.C., Mathias M.L., Collares-Pereira M.J., Viegas-Crespo A.M, Response of antioxidant enzymes in freshwater fish populations (*Leuciscus alburnoides* complex) to inorganic pollutants exposure, The Science of Total Environ. 2001, 280, 153-163.
4. Lopes P.A., Viegas-Crespo A.M., Nunes A.C., Pinheiro T, Marques C., Santos M.C., Mathias M.L., Influence of age, sex and sexual activity on trace element levels and antioxidant enzyme activities in field mice (*Apodemus sylvaticus* and *Mus spretus*), Biol. Trace Elem. Res. (in press).

[1] Instituto Superior de Ciências da Saúde, Monte da Caparica, Portugal
[2] Centro de Estudos de Bioquímica e Fisiologia, Fundação da Faculdade de Ciências, Universidade de Lisboa, Portugal
[3] Centro de Biologia Ambiental, Fundação da Faculdade de Ciências, Universidade de Lisboa, Portugal
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[5] Clínica Universitária de Pneumologia, Associação para a Investigação e Desenvolvimento da Faculdade de Medicina, Universidade de Lisboa, Portugal
[6] CNA, CSIC, Univ. Seville, Spain
[7] Environmental Analytical Chemistry, Chemistry Sector, ITN
[8] INETI, Lisboa, Portugal

# Condensed Matter Physics

*Frederico Gama Carvalho*

The Group's field of research is the development and characterisation of materials, with radiation as a tool to investigate the structure and/or induce structural modifications in samples of the interesting material aggregates. The group activities are two-fold: condensed matter research proper, including the preparation and characterisation of special samples; and instrument development, including design optimisation, construction, installation and operation of equipment and facilities for low-energy neutron scattering and X-ray diffraction. Systems currently under investigation include natural stones, such as topaz, high temperature alloys and organic-inorganic hybrid materials. In both areas of neutron and X-ray for structural studies the Group aims at playing a significant role in the development and implementation of techniques in the country. Developing scientific contacts with national Universities has a high priority in tune with the policy of providing a specialised research infrastructure built around the local facilities available to university researchers.

***Neutron beam facilities***. A Two-Axis neutron Diffractometer equipped with a "banana" multidetector and a rotating chopper Time-of-Flight Diffractometer, are installed in two beam tubes of the RPI research reactor; a Small Angle Neutron Scattering Instrument (SANS) is currently under installation. The operation of the last two instruments is a key point to justify the continued operation of the reactor. The TOF diffractometer is dedicated to the training of students. These experimental facilities can be instrumental in training a school of neutron scattering users in the country that will join the european neutron scattering community in using higher neutron flux sources. Although the RPI neutron flux is sufficient to measure certain classes of samples recourse to other facilities will be essential in many cases. At the same time consideration should be given in due time to an increase of the reactor power and eventually in the long run to the installation of a cold neutron source. In the near future it is foreseen to introduce a section of bent guide in the beam line of the SANS instrument in order to improve the signal to noise ratio at the higher wavelengths.
In 2001 installation of the beam line of the Two-axis Neutron Diffractometer DIDE was completed and first test spectra were recorded showing good agreement with the theoretical predictions made as concerns intensity and resolution. Collaboration of the Laboratoire Léon-Brillouin, CEA/CNRS, Saclay, France, has to be commended. Further investment is required namely in the sample environment in order that the instrument becomes fully operational.
SANS in-pile components were installed following the completion of a detailed report on installation procedures and safety aspects of the spectrometer's operation carried out in collaboration with the Nuclear Instrumentation Group. The Reactor team provided adequate support and advice during the installation of the two instruments.
The design of the first prototype of a neutron Converging Multichannel Collimator was carried out and considerable progress was made towards the solution of the main technical difficulties presented by the collimator's construction, namely the fabrication of the thin neutron absorbing strips that will form the collimating planes of the device. The work is funded by the National Science Foundation and the IAEA.
In the field of neutron scattering collaboration with the Glass and Ceramics Dept. of Aveiro University, Laboratoire Léon Brillouin (Saclay), the Budapest Neutron Center and Sophia University has continued.

***The activity at the MA$^3$T laboratory.*** During 2001, the second year of activity of the High Temperature Materials Laboratory, emphasis was put on the tackling of problems that cannot be solved by conventional diffractometers. Thus, a large part of the research work occurred in collaboration with external Portuguese and foreign research groups in two main fields: electronic structures and high-temperature alloys. Most of this work was centred on single crystalline materials. However, several polycrystalline materials have been studied, namely magnetic multilayer sensors, nano-precipitates in implanted materials, ceramic powders, etc.
The development work at the Hotbird continued both in the software and the hardware fronts. The main development consisted in the addition of a new geometry, the *high-intensity mode* to complement the existing ones: the *double-crystal* and the *triple-axis diffractometer* geometries.
The high-resolution X-ray diffractometer, Hotbird, has been used almost uninterruptedly. Although this has confirmed the reliability of the main hardware parts, this had a clear impact on some of the components. This intense use raised the need for a careful and costly maintenance plan. However, the large hiatus in the availability of financing from the National Science Foundation (FCT) brought serious concerns on the feasibility of the referred maintenance plan. In particular, the evaluation of the last project applications to the FCT, one year ago, have been interrupted. Therefore, if a main failure occurs, the laboratory may suffer a long interruption.

# Condensed Matter Physics

## Research Team

### Researchers

- F.G. CARVALHO, Coordinator Researcher, Group Leader (70%)
- J.F.SALGADO, Coordinator Researcher (15%),
- F.M.A. MARGAÇA, Principal Researcher
- A.N. FALCÃO, Auxiliary Researcher
- A.D. SEQUEIRA, Auxiliary Researcher
- J.S. NEVES, Auxiliary Researcher (20%)

### Students

- N.P. FRANCO, MSc Student, ITN grant
- P. FERREIRA, PhD Student, IST/ITN, FCT grant
- J.L. COSTA, PhD Student, IST/ITN, FCT grant
- J.S. SOUSA, BIC PRAXIS XXI
- M. CARRAPIÇO, BIC POCTI
- D. SILVA, undergraduate student

## Funding

| | $\times 10^3$ PTE |
|---|---|
| Research Projects: | 8069 |
| ITN: | 1400 |
| Other Sources: | 000 |
| **Total:** | **9469** |

## Publications

| | | |
|---|---|---|
| Journals: | 11 | and 4 in press |
| Proceedings: | 2 | and 4 in press |
| Conf. Communications: | 12 | |
| Theses: | 1 | |

# Application of High-Resolution X-ray Diffraction to Study Strain Status in $Si_{1-x}Ge_x/Si_yGe_{1-y}/Si$ (001) Heterostructures

A.D. Sequeira, N. Franco, K.D. Chtcherbatchev[1], N.P. Barradas[2], M. Myronov[3], O.A. Mironov[3], E.H.C. Parker[3]

## Objectives

The aim of the current study was the structural characterisation of p-type modulation doped $Si_{1-x}Ge_x/Si_yGe_{1-y}/Si$ heterostructures using high resolution X-ray diffraction. In particular to study the strain status, thickness and thermal stability of HMOS transistors after thermal annealing at temperatures ranging from 600 to 900ºC.

This work is being carried out in collaboration with Prof. Parker´s group of the University of Warwick.

## Results

The main characterisation tool used was the reciprocal space mapping of symmetric and asymmetric reflections using High-resolution X-ray diffraction. To study the thermal stability and strain relaxation mechanisms in p-type modulation doped $Si_{1-x}Ge_x/Si_{1-y}Ge_y/Si(001)$ heterostructures on virtual substrates. Ex-situ post-growth furnace thermal treatments were done in a N2 ambient at 600, 700 and 750 and 900ºC. It was found that the high temperature annealing leads to the full relaxation of the intermediate part of the virtual substrate (corresponding to the Ge composition range of y=0.17-0.27). Both the uppermost and lowermost parts of the virtual substrate regions with, respectively, $y_{Ge}$=0.27-0.35 and 0.10-0.17, were found to be not fully relaxed even after annealing at 750ºC. The increase of the annealing temperature leads to a broadening of the channel layer with a decreasing of its Ge concentration though remaining fully strained.

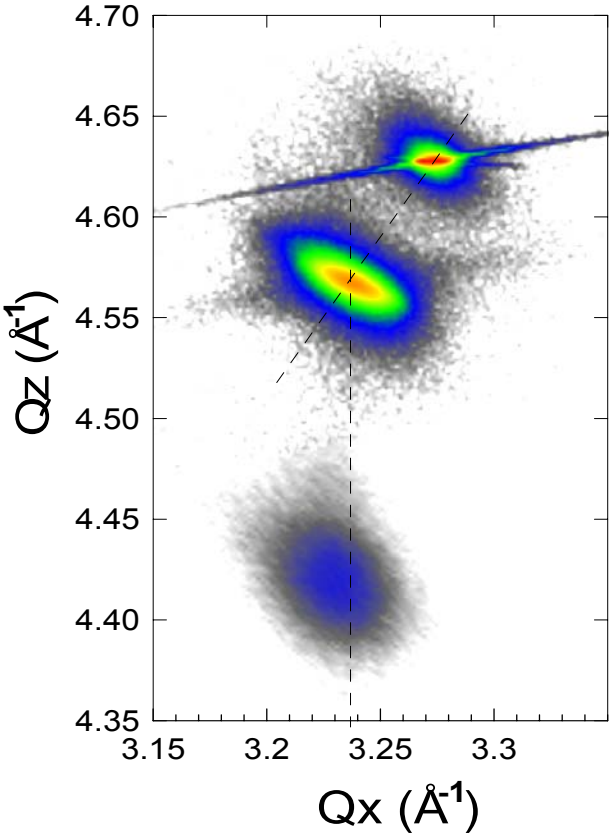


**Fig.** Reciprocal space mapping of the 224 reflection of a heterostructure of $Si_{1-x}Ge_x/Si_yGe_{1-}$

The use of the Hotbird X-ray diffractometer with its powerful rotating anode source and optimised geometry allowed us to reveal and study the very weak intensity scattering from a 4nm thick $Si_{0.2}Ge_{0.8}$ channels.

[1] Moscow State Institute of Steel and Alloys, Russian Federation.
[2] Nuclear Solid State Physics using ion Beams group.
[3] Department of Physics, University of Warwick, Coventry, CV47AL, UK.

## Published, accepted or in press work


1. K.D. Chtcherbatchev, A.D. Sequeira, N. Franco, N. Barradas, M. Myronov, O.A. Mironov, E.H.C. Parker. *Materials Science and Engineering B.* (2002). In press.

2. N.P. Barradas, A.D. Sequeira, N. Franco, M. Myronov, O.A. Mironov, P.J. Phillips, E.H.C. Parker Modern Physics Letters B Vol. 15, Nos. 28 & 29 (2001) 1297.

3. M. Myronov, O.A. Mironov, P.J. Phillips, E.H.C. Parker N.P. Barradas, A.D. Sequeira, N. Franco, E. Alves. *2nd International Conference on Silicon Epitaxy and Heterostructures E-MRS 2001***.** June 4-8 (2001) Strasbourg, France. Poster.

4. K.D. Chtcherbatchev, A.D. Sequeira, N. Franco, N. Barradas, M. Myronov, O.A. Mironov, E.H.C. Parker. *9th International Conference on Defects - Recognition, Imaging and Physics in Semiconductors, DRIP IX*. Rimini, Italy, September 24-28 (2001). Poster.

5. N.P. Barradas, A.D. Sequeira, N. Franco, M. Myronov, O.A. Mironov, P.J. Phillips, E.H.C. Parker. Grazing angle RBS analysis of $Si/Si_{0.7}Ge_{0.3}/Si_{0.2}Ge_{0.8}/Si_{0.7}Ge_{0.3}$ heterostructures. Workshop on Advanced materials produced and analysed with ion beams, Wellington, New Zealand (2001) July 10-12. Oral.

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# Determination of Structural Properties of InGaN

A.D. Sequeira, N.P. Franco, S. Pereira[1], E. Alves[2], K.P. O'Donnell[3]

## Objectives

The main aim of the current project is to study the effect of strain on the optical properties of epitaxial single layers of $In_xGa_{1-x}N$. The main characterisation technique is high-resolution x-ray diffraction.

## Results

***The material.*** Group III nitride epilayers are currently a major topic of research due to their widespread use in light emitting diodes (LED) and laser diodes (LD). Commercial indium gallium nitride (InGaN) LEDs, developed over the last decade, enjoy unrivalled performance in the UV, blue and green spectral regions

***The XRD analysis.*** In this study emphasis has been given to the characterization the structural properties of InGaN structures of low dimensionality in particular multilayers and quantum dots (an example is give in the Fig.). The reciprocal space maping is a technique particularly suited to determine structural parameters of these structures. In particular the strain status of the InGaN layers relative to the GaN substrate. The main problems that have been addressed and published are the following:

1) A new method was developed for the determination of the composition of the InGaN films;
2) Alternative model was developed to explain the appearance of two peaks that has been claimed to be phase separation;
3) An anomalous dependence of the energy gap on the In content was found;

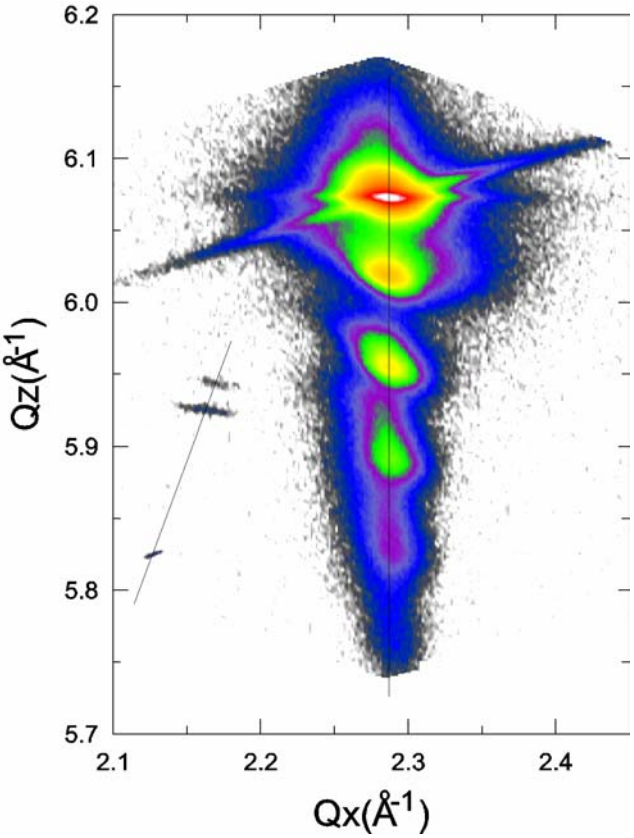


**Fig.** Reciprocal space map of a InGaN superlattice.

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[2] Nuclear Solid State Physics using ion Beams group, ITN
[3] Department of Physics, University of Strathclyde, Glasgow G4 0NG, Scotland, UK.

## Published, accepted or in press work


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3. S. Pereira, M.R. Correia, E. Pereira, K.P. O'Donnell, C. Trager-Cowan, F. Sweeney, E. Alves, A.D. Sequeira, N. Franco and I. M. Watson. Depth Resolved Studies of Indium Content and Strain in InGaN Layers. *Phys. Stat. Sol. (B)* **228, 1** (2001) 59.

4. E. Alves, S. Pereira, M. R. Correia, E. Pereira, A.D. Sequeira, N. Franco and K. P. O'Donnell, C. Trager-Cowan. Strain Relaxation and Compositional Analysis of InGaN/GaN layers. *Nuclear Instruments and Methods B.* In press.

5. S. Pereira, M.R. Correia, E. Pereira, K.P. O'Donnell, R.W. Martin, M.E. White, E. Alves, A.D. Sequeira, N. Franco. Splitting of X-ray diffraction (XRD) and photoluminescence (PL) peaks in InGaN/GaN layers. *Mat. Sci. Eng. (B).* In press.

6. S. Pereira, M.R. Correia, E. Pereira, C. Trager-Cowan, F. Sweeney, P.R. Edwards, K.P. O'Donnell, E. Alves, A.D. Sequeira, N. Franco. Strain and Compositional Analysis of InGaN/GaN Layers in *GaN and Related Alloys – 2000*, Edited by U. Mishra, M.S. Shur, C.M. Wetzel, B. Gil, K. Kishino (*Mater. Res. Soc. Proc.* **639**, Pittsburgh, PA, 2001) G 3.52, 1.

7. E. Alves, S. Pereira, M.R. Correia, E. Pereira, A.D. Sequeira, N. Franco and K.P. O'Donnell, C. Trager-Cowan. *15th International Conference on Ion Beam Analysis, IBA-15.* 15-20 July, Cairns, Australia (2001). Oral presentation.

8. S. Pereira, M.R. Correia, E. Pereira, K.P. O'Donnell, C. Trager-Cowan, F. Sweeney, E. Alves, A.D. Sequeira, N. Franco and I. M. Watson. *The Fourth International Conference on Nitride Semiconductors ICNS-4.* July 16-20, Denver, Colorado USA (2001). Poster.

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# Study of Single-Crystalline MCrAlY Coatings Produced by Laser Cladding

A.D. Sequeira, P. Ferreira[1], R.C. da Silva[2] and R. Vilar[1]

## Objectives

The aim of this project is to produce and characterise CoNiCrAlY single-crystalline coatings on single crystalline superalloys with low porosity and good oxidation resistance at high temperature, using laser cladding. It is very importance to produce coatings free of defects (pores, inclusions) since these defects are associated with crack initiation and accelerate thermal degradation of the coating and subsequently the substrate.

## Results

Despite the improvements brought by the introduction of new alloys and the use of single crystalline materials for turbine blades and vanes, it is impossible to find alloy compositions that
allow the required mechanical and corrosion properties to be simultaneously achieved. As a result, turbine blades must be coated with a material with outstanding oxidation and corrosion resistance, like MCrAlY-type alloys, in order to achieve the required component lifetime.

The deposition of single crystalline MCrAlY coatings on single crystalline turbine blade materials is demonstrated by the authors in the present project. Here one reports on the structure and properties of single crystalline laser cladded coatings. Detailed microstructural characterization and X-ray diffraction experiments were performed in order to establish orientation relationships between the substrate material and the coating material.
Experimental results show that the coating structure develops epitaxially from the substrate and inherits its crystal orientation. The single crystalline character is preserved even when several layers of MCrAlY are overlapped.

This work showed that it is possible to produce single crystalline coatings of a MCrAlY alloy on single crystalline substrates by means of laser cladding. The coating preserves the orientation of the substrate and its crystalline quality is better or at least comparable to the crystalline quality of the substrate.

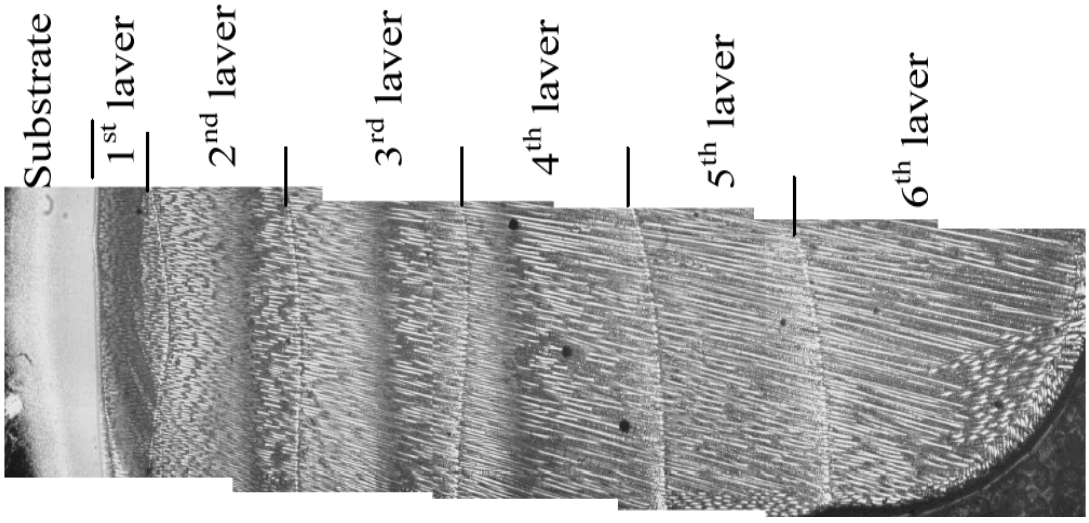


**Fig.** Result of a multiple laser deposition where the whole lenght is single crystalline.

## Published, accepted or in press work

1. P.N. Ferreira, R. Vilar, J.P. Pina, R.C. da Silva and A.D. Sequeira. MCrAlY Coatings Deposited on Single-crystal High-temperature Alloys by Laser Cladding. Proceedings of *ICALEO 2001 – 20th International Congress of Applications of Lasers and Electro-optics*, Laser Institute of America (2001) 521.

[1] IST, Av. Rovisco Pais, 1096 Lisboa Codex, Portugal.
[2] Nuclear Solid State Physics using ion Beams group, ITN

# Strain Development on Hf, In, Ca, Fe and Mn Implanted GaN and its Evolution upon Annealing

A.D. Sequeira, E. Alves[1], N. Franco, K. Lorenz, R. Vianden[2], N.A. Sobolev[3], M.C. Carmo[3]

## Objectives

The aim of the current project is to determine the strain and damage produced on GaN during the implantation process of several species (Ca, Fe, Hf, In, etc.), the dependence on implanted dose and its evolution under different annealing conditions.

## Results

After the characterisation of several implanted species (Mn, Ca, Fe) the current studies are centred in Hf and In. These characterisation of the latter elements is being made in collaboration with Bonn University where complementary studies using perturbed angular correlations (PAC) are being performed.

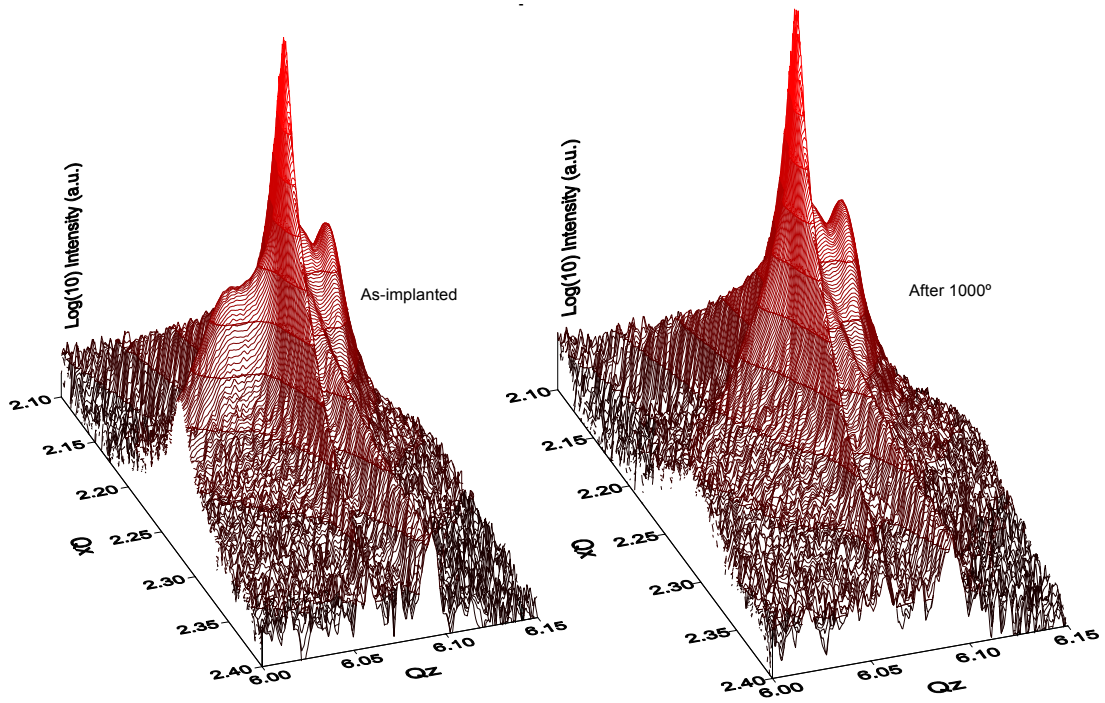


**Fig.** Reciprocal space maps of Hf implanted GaN. After implantation (left) and after annealing at 1000ºC. Note the recovery of the damage (left hand peak produced by the implantation) after the annealing.

## Published, accepted or in press work

1. C. Liu, E. Alves, A.D. Sequeira, N. Franco, M.F. da Silva and J.C. Soares. Fe ion implantation in GaN: Damage, annealing and lattice site location. *J. Appl. Phys.* **90**, 1 (2001) 81.

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3. C. Liu, A. Wenzel, B. Rauschenbach, E. Alves, A.D. Sequeira, N. Franco, M.F. da Silva, J.C. Soares, X.J. Fan. Amorphization of GaN by ion implantation. *Nuclear Instruments and Methods* Vols. **178** (2001) 200.

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[2] Institute für Strahlen- und Kernphysik, Universität Bonn, Nuβallee 14-16, 53115 Bonn, Germany.
[3] Department of Physics, University of Aveiro, 3810-193 Aveiro, Portugal.

# Structural analysis of Mesoporous Materials - MCM-41

A.D. Sequeira, M.M.L. Ribeiro Carrott[1], A.J. Estêvão Candeias[1], P.J.M. Carrott[1], P.I. Ravikovitch[2] and A.V. Neimark[2]

## Objectives

MCM-41 (Mobile Crystalline Material) is a silicate. It is ordered to some degree, so that there are arrays of non intersecting hexagonal channels. By changing the length of the template molecule, the width of the channels can be controlled to be within 2 to 10 nm. The walls of the channels are amorphous $SiO_2$.
The aim of the current project is to correlate the adsorption properties of MCM-41 amorphous materials with the dimensions of the hexagonal lattice structure into which the pores organize themselves.

## Results

The determination of the pore structure was performed using grazing incidence X-ray diffraction. The measurements are delicate due to the small scattering angles of the main peaks which are very close to the incident beam. Analysis of the results indicates that the density of the nitrogen in the pores is higher than that of the normal liquid. In addition, the results show that in order to calculate the surface area of the pore walls, which is needed in order to calculate the hydraulic pore width, a value for the cross sectional area of the nitrogen molecule lower than the normal value must also be used.

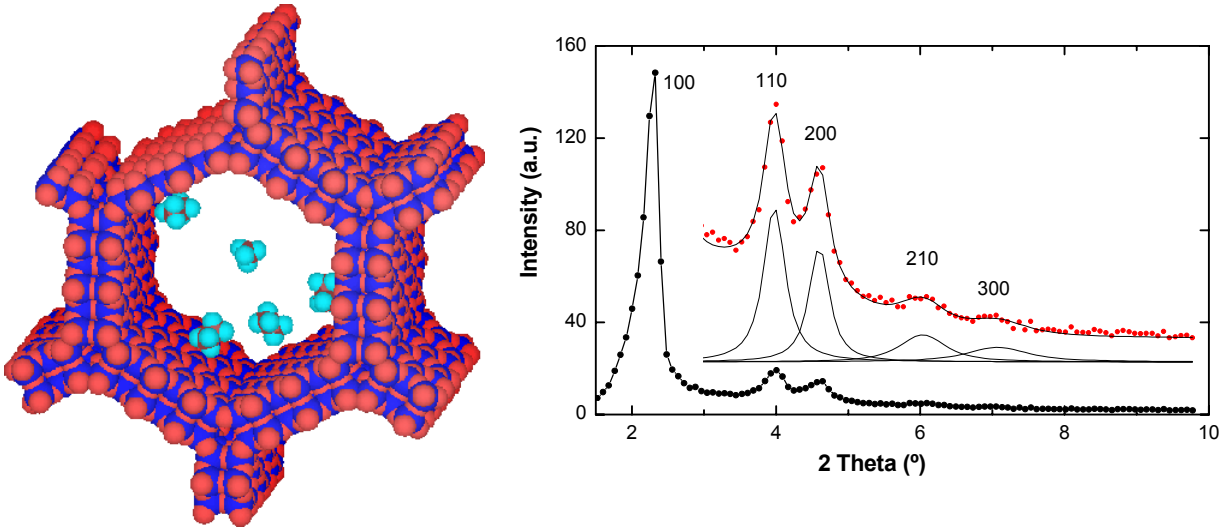



**Fig.** Model of the MCM-41 molecule(left) and XRD spectrum of a MCM-41 sample (right).

## Published, accepted or in press work

1. M.M.L. Ribeiro Carrott, A.J. Estêvão Candeias, P.J.M. Carrott, P.I. Ravikovitch and A.V. Neimark and A.D. Sequeira. Comparison of the adsorption of nitrogen with neopentane, n-hexane, benzene and methanol to evaluate pore sizes in silica grades of MCM-41. *Langmuir* (2001).

[1] Department of Chemistry, University of Évora, Colégio Luís António Verney, Évora, Portugal
[2] TRI - Princeton, 601 Prospect Avenue, Princeton, New Jersey, USA..

# Software and Hardware Development at Hotbird

N.P. Franco, J.L.Costa[1] and A.D. Sequeira

## Objectives

The development work is being made in two fronts: specific hardware upgrades and the continuous updating of software used for data collection and data analysis.

## Results

**Software.** New tools for data analysis have been developed in particular the program 2D spectra adjust for the analysis of reciprocal space maps (see Figure).

A second program developed this year is a dedicated application for the remote control of the high-temperature chamber.

**Hardware.** During the current year a new alternative configuration has been added to the Hotbird, the high-intensity geometry. It uses a direct beam and the wavelength selection is made by a crystal analyser. The third generation of the high-temperature chamber is also being finalized.

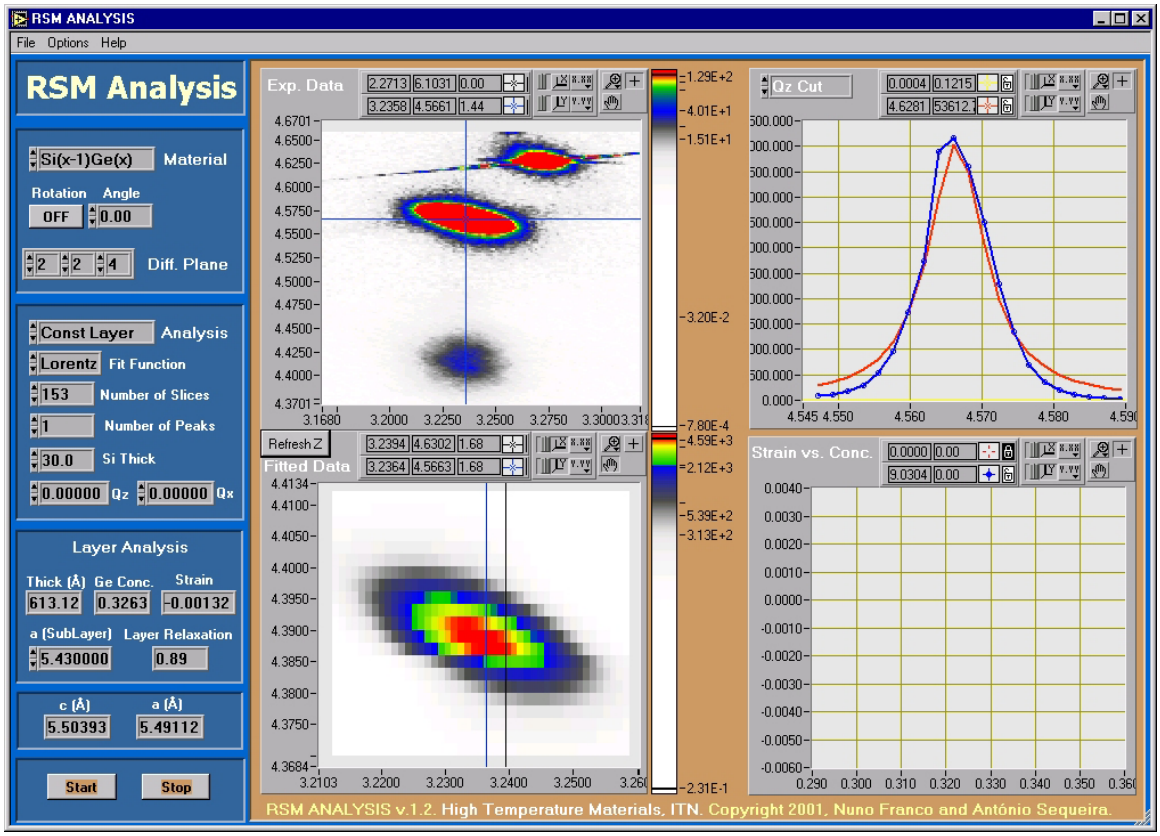



**Fig.** – User interface of the *RSM Analysis* application for data analysis of 2D reciprocal space maps.

## Published, accepted or in press work

1. A.D. Sequeira, N. Franco and J. Neves. High-Resolution and High-Temperature Double-Crystal X-ray Diffractometer for in-situ Studies - the Hotbird. *Mat. Sci. Forum* Vols**. 378-381** (2001) 206.

2. N. Franco and A.D. Sequeira. Software Package for the Remote Control and Data Acquisition from a High-temperature X-ray Diffractometer The Hotbird. *Mat. Sci. Forum* Vols. **378-381** (2001) 212.

[1] IST, Av. Rovisco Pais, 1096 Lisboa Codex, Portugal.

# Development of a Neutron Converging Multichannel Collimator

A.N. Falcão, F.M.A. Margaça, J.F. Salgado and F.G. Carvalho

## Objectives

It has been shown that the use of a converging multichannel collimator (CMC) can improve considerably the performance of existing small angle scattering facilities. The objective of this work is to design, construct and test a variable geometry CMC.

## Results

The general concept of the variable geometry CMC was introduced. Variation of geometry is made possible by replacing continuous walls by interlaced wire or ribon planes. The ability of such a device to prevent cross-talk between neighbouring elementary channels was established by means of a Monte-Carlo computer simulation study. The device performance measured by the balance intensity/resolution, was assessed in the simulation work. The results obtained confirmed the theoretical predictions, and revealed interesting resolution effects that will be reported in a paper shortly to be submitted for publication.

Practical considerations led to choosing the thin ribbon version for the construction of the collimator walls. The materials and the experimental procedure required to obtain a ribbon with the required mechanical and neutron absorption properties were developed. Experimental tests confirmed the good quality of the ribbon samples fabricated. The working life time of the ribbon was estimated based on a simulation study of the dose deposited, and an irradiation test in a $^{60}$Co irradiation unit where severe working conditions were simulated. The design of a first prototype was concluded. The construction of the prototype is presently under way.

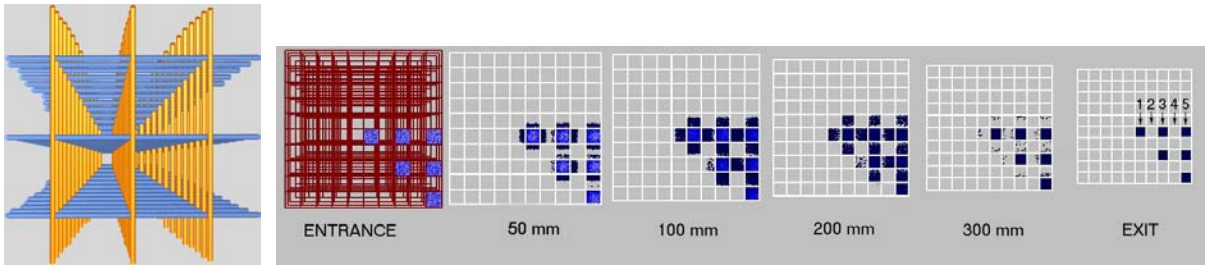



**Figure 1**. (left) - Schematic view of a converging CMC with interlaced wire planes. (right) - Results of the Monte-Carlo simulation: neutrons were injected in 6 representative channels of the CMC device. Their spatial distribution is sampled at different planes perpendicular to the beam axis placed along the CMC. At the exit, note that the entrance pattern is exactly reproduced guaranteeing the absence of cross-talk.

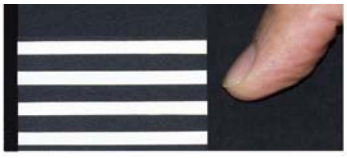


**Figure 2.** Picture of the thin ribbon developed. Each element is 3.5 mm wide and 0.2 mm thick.

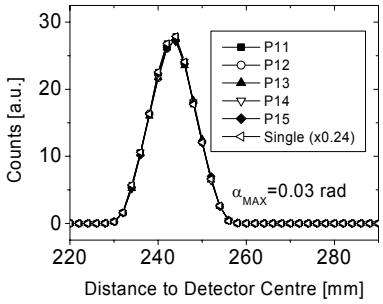



**Figure 3.** Peaks obtained from neutrons transmitted by channels 1 to 5 (Fig. 1) and by a single channel collimator normalised to the same integrated intensity when the maximum incoming beam divergence is $\alpha_{MAX}$ =0.03 rad

## Published, accepted or in press work

1. A. N. Falcão, F.M.A. Margaça and F.G. Carvalho, The use of multichannel collimation in small angle neutron scattering: a computer simulation study, *Appl. Phys. A* (2002) in press.

# Preparation and Characterisation of Organic-inorganic Hybrid Materials

*A.N. Falcão, J.S. Sousa, F.M.A. Margaça, F.G. Carvalho, I.M.M.Salvado[1], J. Teixeira[2] and M. Misheva[3]*

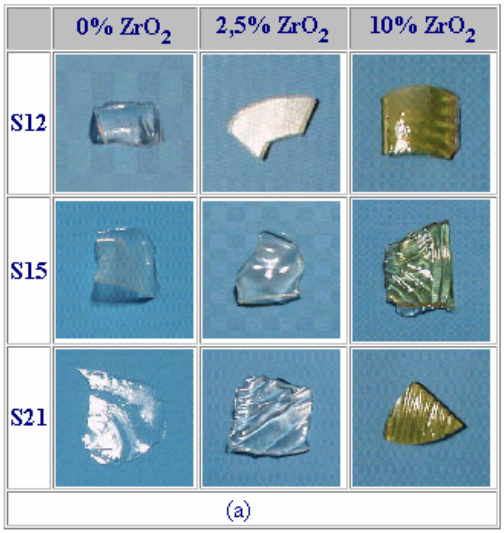
## Objectives

Preparation and characterisation of hybrid materials with different properties by changing polymer concentrations and molecular weights, inorganic composition, processing conditions and using new preparation methods.

## Results

Hybrid materials, consisting of PDMS, chemically bound to (100-x)$SiO_2$.x$MO_2$ (M=Ti,Zr and x=10 mol%) were prepared by sol-gel as well as by gamma irradiation of the precursors.

The former, referred to as gel hybrids were prepared from PDMS of average molecular weight, $M_w$ = 400-700, 1500-2000 and 4200 denoted S12, S15 and S21, respectively (Figure 1).



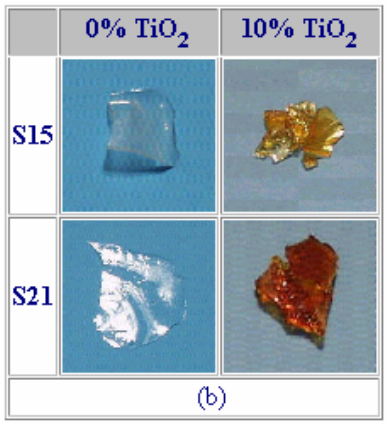




Fig. 1 - Samples of hybrids with a range of compositions prepared by sol-gel.

Irradiated hybrids were prepared by thoroughly mixing the inorganic precursors with PDMS of average molecular weight $M_w$ = 36 000, and irradiating the samples up to a dose of 700 kGy at the ITN $^{60}$Co Gamma Irradiation Facility. Photographs of irradiated samples are shown below (Figure 2).

Silica based hybrid materials, produced by gamma irradiation or by sol-gel, are observed to be transparent and homogeneous at the macroscopic scale.

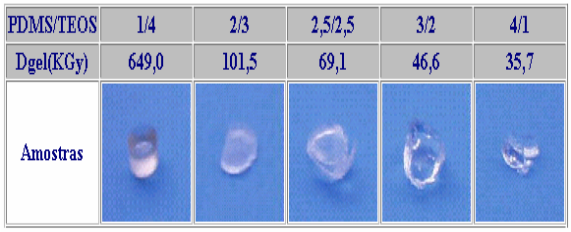



**Fig. 2** - Samples prepared by gamma irradiation at different doses and organic-inorganic component mass ratios.

Hybrids prepared by irradiation are able to swell in a good solvent of the polymer, their microscopic structure was found to be formed by oxide particles interconnected by polymer structures.

Hybrids prepared by the sol-gel process show no swelling, their microscopic structure consisting of an open mass fractal oxide network, where the well distributed extended polymer is linked.

In both hybrids the addition of zirconia alters the microscopic structure of the sample.

## Published, accepted or in press work


- 1. A. N. Falcão, J. Santos Sousa, F.M.A. Margaça, L. M. Ferreira, F.G. Carvalho, I.M. Miranda Salvado and J. Teixeira, Investigation of organic-inorganic materials prepared by irradiation, *J. Sol-Gel Sci. and Tech.* (2002) in press.
- 2. A. N. Falcão, J. Santos Sousa, F.M.A. Margaça, F.G. Carvalho, I.M. Miranda Salvado and J. Teixeira, SANS Study of zirconia-silica sol-gel prepared hybrid materials, *J. Sol-Gel Sci. and Tech.* (2002) in press.
- 3. M. Misheva, N. Djourelov, F.M.A. Margaça and I.M. Miranda Salvado, Positronium study of sol-gel prepared SiO2: influence of pH, J. *Non-Cryst. Solids* **279** (2001) 196.


[1]Glass and Ceramics Engineering Dept., UIMC, University of Aveiro, 3810 Aveiro, Portugal
[2]Léon Brillouin Laboratory (CNRS/CEA), C.E. Saclay, 91191 Gif-sur-Yvette Cedex, France
[3]Faculty of Physics, Sofia University, 5 J. Bourchier Boulevard, 1126 Sofia, Bulgaria

# Instruments for Neutron Scattering at the Portuguese Research Reactor

A.N. Falcão, F.M.A. Margaça, J.F. Salgado and F.G. Carvalho

## Objectives

To provide a specialised research infrastructure built around the local research reactor that is available to university researchers and other external users, furthers the reactor utilisation and contributes to the development and implementation of neutron scattering technique in the country.

## Results

Installation of the beam line of the Two-axis Diffractometer DIDE was completed. The monochromator equipped with a set of Grafite filters was placed in position and tested together with the remote control electronics previously developed. The multidetector and respective electronics were tested in collaboration with the electronics group of the *Laboratoire Léon-Brillouin*, CEA/CNRS, Saclay, France. The first test spectra were recorded. The results obtained reproduced essentially the theoretical predictions that had been made, and were quite promising as to the possibility of future work. Thourough tests of the performance were carried out showing that additional filtering of fast neutrons is not necessary, but the filtering of higher order components of the monochromated beam has to be improved. The essential problem identified during the test phase was the high level of electromagnetic interference observed.

It is expected that the instrument will be commissioned in the beginning of 2002.

A light chopper to be used for the determination of the energy distribution of a thermal neutron beam was developed. Pulse width and frequence are set by remote control. The apparatus is, presently under construction.

The new detector bank that equips the time-of-flight diffractometer ETV was tested, and the acquisition software upgraded. The instrument was used for training and educational purposes in general.

As concerns the Small Angle Neutron Scattering Instrument, EPA, the second part of the report for the safety analysis was delivered in May. Permission to start the installation work was granted in October. Installation work started in November. The installation of the in-pile components in beam tubes E2 and D2 were completed in December. The installation work of the out-of-pile equipment is in progress. It is foreseen to proceed with the installation and testing of different components and of the spectrometer as a whole so that the instrument is commissioned during the first half of 2002.

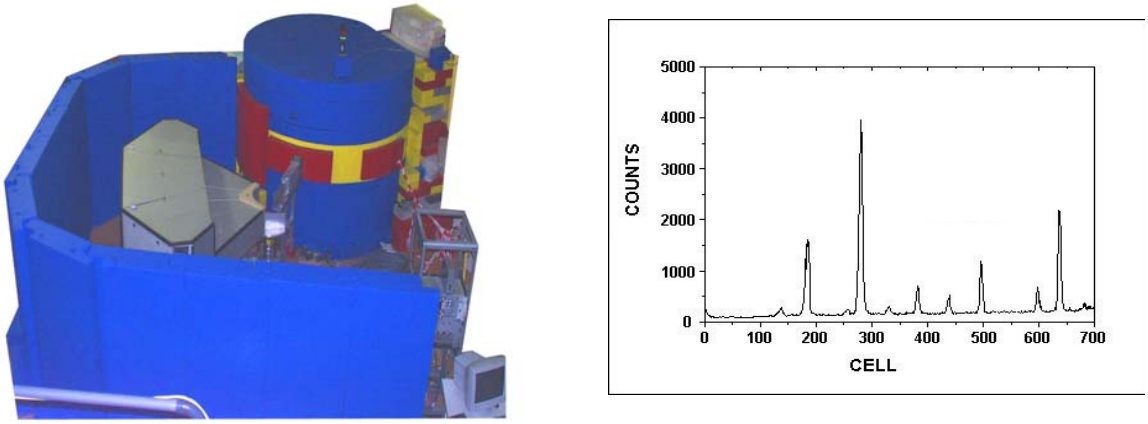



**Figure 1** – View of the two-axis diffractometer DIDE, and spectrum recorded during the test phase.

# Nuclear Instruments and Methods

*José Salgado*

The strategy of the group involves activities in two different lines: *Computational Physics*, and *Instrumentation and Technical Assistance*. Numerical modelling represents an important tool to fulfil the objectives of the group and two domains of application are presently covered: *Calculation of radiation fields* and *Modelling of cold plasmas for material processing applications*.

The group has assembled two Beowulf cluster for parallel computation - a small cluster built with 16 486/66 processors used for demonstration, testing and development of parallel software and a cluster using 3 Pentium III processors, routinely used for Monte Carlo simulations.

**Calculation of radiation fields**

The calculation of radiation fields has a broad range of applications, such as medical physics, material processing, radiosterilisation, reactor irradiation and radiation transport in superconductors. Currently the group is involved in three of these areas:

*Medical physics*
Characterisation of an electron linear accelerator was carried out through the use of MCNP code:
1. Monte Carlo simulation of narrow photon beams used in radiosurgery allowing the best understanding of local energy deposition besides of the lack of electron equilibrium.
2. Monte Carlo calculation of relevant dosimetric parameters for non-regular fields induced by cerrobend inserts placed into standard electron applicators.

*Radiosterilisation*
1. The study of the irradiator geometry of the Portuguese Gamma Irradiation Facility, using the MCNP code.
2. The study of the influence of the radiation on the properties of disposable surgical clothing. Three types of materials (nonwoven, laminate and polyethylene), two types of radiation (electron beam and gamma radiation) and three sets of dose values –25, 80 and 160 kGy - were considered**.**

*Reactor physics*
Neutron resonance self-shielding factors in different materials (Au, Co, Cu, In, Mn, Re) and different geometries (foils, wires, spheres and) have been calculated.
It was shown that an adimensional variable, z, can be introduced ($z=\Sigma_{tot}(E_{res}).x.(\Gamma_\gamma/\Gamma)^{1/2}$), which converts the dependence of the resonance self-shielding factor on physical and nuclear parameters into an "universal" curve, valid for all materials.

*Other applications*
A study of the response of geometrically-metastable type I super conducting strips of Re, Al, Sn and Pb to low energy laboratory electron and X-ray irradiations, towards an improved understanding of the basic physics underlying the strip response necessary to both the development of new particle detection devices and in the analyses of current results on the beta decay of $^{187}$Re.
New quasi-monoenergetic neutron calibrations of the SIMPLE detector at RPI using filtered neutron beams of energies 24.3±2, 55±2 and 144±24 keV, with the goal of exploring the larger than calculated response noted at low temperatures in previous neutron irradiations of a SIMPLE module performed at the CERN using $^{252}$Cf.

**Electrical Discharges on Environment and Material Processing Applications**

Cold plasmas have a significant impact in material processing and environment applications. The activities on these areas have been divided between numerical simulation work and experimental research.
*Simulation work:* A new code for the solution of electron Boltzmann equation in cold plasmas and molecular gases has been completed. The application of a genetic algorithm to cross section fitting has been successful. The simulation of a glow discharge in $N_2$ with a carbon cathode has progressed.

*Experimental work:* A laboratory plasma discharge system for gas cleaning has been projected and is under construction. A gas analysis system for atmospheric pressure sampling has been projected and is been assembled. A Langmuir electrostatic probe has been built.

**Instrumentation and Technical Assistance**

The main objectives of this activity are the technological support for internal groups, including specific design for nuclear instruments, and technical assistance to industrial companies.
Equipment has been designed for the *DIDE* and *SANS* facilities.
A new series of the Personal Dosimeter RAD X100, with some improvements as well as an optimised Portable Gamma Level indicator have been designed.
The technical assistance takes mainly the forms of specialized consultant engineering advice, installation of nuclear gauges, including calibration maintenance and repair and recharging of gauges with imported sources.
The Group is also involved in the n_TOF collaboration, a consortium of 40 laboratories in Europe and USA. The foreseen participation of the Group is: (a) Monte Carlo simulation of the experimental set-up, (b) development of C6D6 detectors.

# Nuclear Instruments and Methods

## Research Team

### Researchers

- J. SALGADO, Coord. Researcher, Group Leader (70%)
- F. G. CARVALHO, Coord. Researcher (10%)
- C. OLIVEIRA, Principal Researcher
- J. MANTEIGAS, Auxialiary Researcher (70%)
- I. F. GONÇALVES, Auxialiary Researcher
- J. NEVES, Auxialiary Researcher (80%)
- C. CRUZ, Auxialiary Researcher
- N. PINHÃO, Auxialiary Researcher

### Students

- A. CHAVES, PhD Student
- M.R.V. Martins, PhD Student
- J. AMBRÓSIO, MSc Student
- M.S. SILVA, undergraduate Student
- A. GONÇALVES, (grant Sociedade Ponto Verde)

### Technicians

- T. JESUS, laboratory technician
- N. INÁCIO, laboratory technician (85%)

## Funding

| | $\times 10^3$ PTE |
|---|---|
| Research Projects: | 766 |
| Services: | 2615 |
| **Total:** | **3381** |

## Publications

| | |
|---|---|
| Journals: | 5 and 4 in press |
| Proceedings: | 4 and 3 in press |
| Conf. Communications: | 8 |
| Internal reports: | 1 |
| Other publications: | 4 |
| Theses: | 2 graduation |

# Calculation of neutron resonance self-shielding factors in wires, foils, spheres and cylinders of different materials

I. F. Gonçalves, J. Salgado, E. Martinho[1]

## Objectives

The cross sections of most materials used in neutron dosimetry or for radionuclide production exhibit resonances in the epithermal region of the reactor neutron spectrum. The aims of this work are (a) to calculate the neutron resonance self-shielding factor in different materials and different geometries, (b) to interpret its dependence on the physical and nuclear parameters of the studied materials.

## Results

Simulation studies were carried out with the MCNP code to calculate the neutron resonance self-shielding factor in different materials (Au, Co, Cu, In, Mn, Re) and different geometries (foils, wires, spheres and cylinders) [1, 2, 3, 4].

The resonance self-shielding factor of a nuclide depends on material density and dimensions, atomic mass, natural abundance, resonance cross section and resonance widths for neutron scattering, $\Gamma_n$ and radiative capture, $\Gamma_\gamma$.

The energy dependent self-shielding factors, $G_{res}(E)$, of materials with high capture cross sections have different behaviour than those of materials with high scattering cross-sections. In the first case, $G_{res}(E)<1$ for the whole energy interval; in the second case, $G_{res}(E)<1$ for $E<E_{res}$ because neutrons entering with energy $E_{res}$ can suffer one or more scattering interactions before being captured at lower energy.

It was shown that an adimensional variable, $z$, can be introduced ($z=\Sigma_{tot}(E_{res}).x.(\Gamma_\gamma/\Gamma)^{1/2}$), where x is equal to $R$ for spheres, $1.5t$ for foils and $2R$ for wires), which converts the dependence of the resonance self-shielding factor on physical and nuclear parameters into an "universal" curve, valid for all materials. The figure shows a comparison of the "universal" curve with experimental values obtained by different authors.

## Published, accepted or in press work


1. I. F. Gonçalves, E. Martinho, J. Salgado, Monte Carlo calculation of resonance self-shielding factors for epithermal neutron spectra, *Radiation Physics and Chemistry*, 61 (2001) 461-462.
2. I. F. Gonçalves, E. Martinho, J. Salgado, Monte Carlo calculation of epithermal neutron resonance self-shielding factors in wires for different materials, *Applied Radiation and Isotopes*, 55 (2001) 447-451.
3. I. F. Gonçalves, E. Martinho, J. Salgado, Monte Carlo calculation of epithermal neutron resonance self-shielding factors in foils for different materials, *Applied Radiation and Isotopes (in press).*
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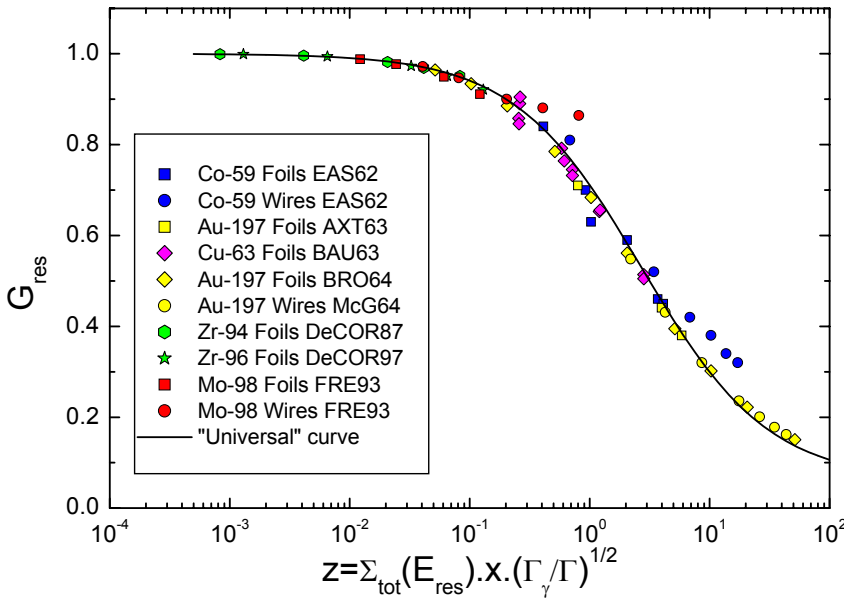


Comparison of the universal curve with experimental points

[1]Reactor, ITN, Sacavém, Portugal

# Monte Carlo optimisation of the irradiator geometry. The impact of gamma and electron beam irradiations over the properties of disposable surgical clothing

C. Oliveira, J. Salgado, I.F. Gonçalves, L.M. Ferreira, M.H. Casimiro, M.E. Cabeço Silva[2], M.J. Marques Abreu[2]and A. Coelho[3]

## Objectives

1. The study of the irradiator geometry of the Portuguese Gamma Irradiation Facility, using the MCNP code.
2. The study of the influence of the radiation on the properties of disposable surgical clothing. Three types of based material (nonwoven, laminate and polyethylene), two types of radiation (Electron Beam and Gamma Radiation) and three set of dose values –25, 80 and 160 kGy - were considered.

## Results

1. Eight different source rack configurations were simulated. The number of cobalt sources that were considered varies between 156 and 296 and the activity between 2.71E15 Bq and 5.14E15 Bq (reported to October 1999). Routine and non-routine irradiations have been studied.
The main conclusions for the routine irradiations are: *i)* the dose uniformity doesn't depend on source configuration but only on products density; *ii)* the average dose rate depends linearly on source rack activity and no saturation effect is observable (Fig.1). For non-routine irradiations the dose uniformity varies significantly, in the inner row, with product density and source configuration. For the outer row these effects are observed to be less important.

2. In order to study of the influence of the radiation on the properties of disposable surgical clothing, three types of based material (nonwoven, laminate and polyethylene), two types of radiation (Electron Beam and Gamma Radiation) and three absorbed doses – 25kGy, 80 kGy and 170 kGy have been delivery to the products. In order to use the Differential Scanning Calorimetry (DSC) and Thermogravimetry (TGA) techniques the products were irradiate in a range from 0 to 400 kGy. The DSC and TGA techniques show that a nonwoven and a laminate textiles maintain a good molecular cohesion, not showing high levels of degradation, for gamma radiation dose values lower than 100 kGy and 200 kGy in nonwoven and in laminate materials, respetively.
The tensile strength and the elongation decrease slowly for the nonwoven textile and decrease faster for the laminate textile for 25 and 80 kGy absorbed dose. The study of the influence of the 160 kGy is in progress.

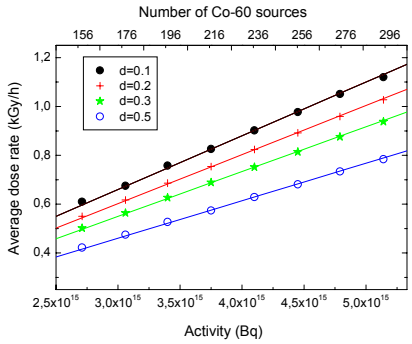


Fig. 1: Average dose rate as a function of source rack activity for the four different densities.

## Published, accepted or in press work

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2. C. OLIVEIRA, L-M. FERREIRA, J. SALGADO. Monte Carlo studies of the Portuguese gamma irradiation facility. The irradiator geometry and its influence on process parameters, MC2000 – "Proceedings MC 2000". 497, Ed. Springer Verlag, (2001)
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4. L.M. FERREIRA, M.H. CASIMIRO, C. OLIVEIRA, M.E. CABEÇO SILVA, M.J. MARQUES ABREU AND A. COELHO. Thermal Analysis Evaluation of Mechanical Properties Changes Promoted by Gamma Radiation in Surgical Polymeric Textiles". NIM-B (in press).

[1] Eng.Textil Department, University of Minho, Portugal
2 Fapomed. Felgueiras.

# Monte Carlo Simulation of Radiosurgery Photons and Electron Beams

A. Chaves[1], J. Ambrósio, M.C. Lopes[1], C. Alves[1], C. Oliveira, L. Peralta[2], P. Rodrigues[2] and A. Trindade[2]

## Objectives

Characterisation of an electron linear accelerator through the use of MCNP code.

1. Monte Carlo simulation of narrow photon beams used in radiosurgery allowing the best understanding of local energy deposition besides of the lack of electron equilibrium.

2. Monte Carlo simulation of electron beams. Calculus of relevant dosimetric parameters for non-regular fields induced by cerrobend inserts placed into standard electron applicators.

## Results

1. A very good agreement was observed between calculated and measured depth dose curves for all secondary collimators used in radiosurgery. Also in the case of output factors and off axis ratios the agreement between the measurements and the calculation is very good (differences less than 2%). Meanwhile a new approach of simulation is being developed. The new method consists in mixing full Monte Carlo simulations with some parameterisations of the sources in order to reduce the running time of the simulation. The new phase space distribution calculated to reduce the fluctuations would be parameterised. This phase space distribution is composed of various distributions, one for each element of the accelerator (photons and electrons). These different sources of particles must be perfectly characterized: contribution to the total phase space distribution, spatial distribution, spectra, and origin of the particles. Once all the sources will be modelled, a particle will be sampled from these distributions and a full Monte Carlo simulation will proceed. Figure 1 shows the example of the spatial distribution of the photons produced in the target that reached the scoring plane.

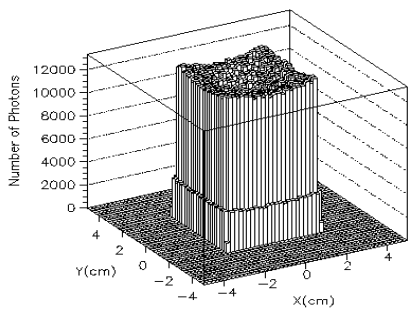


**Fig. 1**. Spatial distribution of photons on the scoring plan originated in the target.

2. Concerning the electron beams two kind of irregular fields have already been studied: a non-centred rectangular 8x4 cm2 frame and a peanut-shaped frame. Relevant data have been analysed and compared with available experimental data and DDP and X- and Y- profile curves have been plotted. Owing to the complexity of MCNP input specifications, a computer program (FRAME) has also been developed and fine tuned in order to automatically generate plane and cell input specifications for MCNP. At the output the end-user obtains the MCNP input specifications corresponding to the designed shape.

Some specific frames have been designed with the program and calculated data compared with experimental dose results for 10 MeV electron energy. The experimental and simulated results reveal a good agreement.

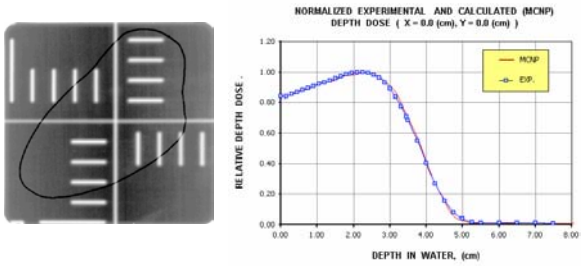


**Fig. 2**. The relative depth dose (experimental and simulated) for the peanut-shaped frame.

## Published, accepted or in press work

1. A. TRINDADE, P. RODRIGUES, C. ALVES, A. CHAVES, M.C. LOPES, C. OLIVEIRA, L. PERALTA. Monte Carlo Simulations of electron beams for radiotherapy – EGS4, MCNP4b and Geant3 intercomparison". MC2000 – "Proceedings MC 2000". 311, Ed. Springer Verlag, (2001)

2. C. ALVES, M.C. LOPES, A. CHAVES, L. PERALTA, P. RODRIGUES, A. TRINDADE, C. OLIVEIRA. Buildup measurements and calculations for photon beams – search of the effective point of measurement for pinpoint and Markus chambers". Physica Medica. Proceedings MeRPE-2000.

[1] CROC-Centro Regional de Oncologia de Coimbra do IPOFG

[2] LIP-Laboratório de Instrumentação e Física Experimental de Partículas. Lisboa

# Geometrically-metastable type I superconducting strips and neutron calibration of the SIMPLE detector

T.A. Girard[1], M.R. Gomes[1], P. Valko[1], J.G. Marques[2], C. Oliveira F. Giuliani[1], J. I. Collar[3], T. Morlat[3], D. Limagne[3] and G. Waysand[3]

## Objectives

1. A study of the response of geometrically-metastable type I superconducting strips of Re, Al, Sn and Pb to low energy laboratory electron and X-ray irradiations, towards an improved understanding of the basic physics underlying the strip response necessary to both the development of new particle detection devices and in the analyses of current results on the beta decay of $^{187}$Re.

2. New quasi-monoenergetic neutron calibrations of the SIMPLE detector at RPI using filtered neutron beams of energies 24.3±2, 55±2 and 144±24 keV, with the goal of exploring the larger than calculated response noted at low temperatures in previous neutron irradiations of a SIMPLE module performed at the CERN using $^{252}$Cf.

## Results

1. Irradiation of a Sn strip by the decay electrons of $^{109}$Cd was attempted several times. The last one, a 50 kBq source was evaporated into a linear groove in a Cu block. Data was obtained with strips of 12.5, 25 and 50 μm thickness, operated at ~300 mK, obtained with field ramp rates of 50 gauss/s.
   A part of the experimental results is shown in Fig. 1 and clearly evidences structure.

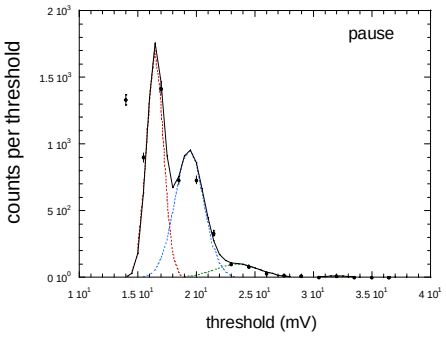


**Fig. 1**. Results of $^{109}$Cd irradiation of a 25 μm tin strip.

A Gaussian fitting is indicated, although the data remains under analyses; a linear calibration assuming the low intensity upper two peaks to correspond to the conversion electrons yields energies of ~22 and ~27 keV for the two lower peaks, consistent with an MCNP simulation which indicates all four peaks in the energy deposition spectrum.

2. The first neutron calibrations of the SIMPLE detector were performed at RPI using filtered beams of 24, 55 and 144 keV. The filter modules consisted of:
   24 keV: Fe (40 cm) + Al (20 cm) + S (20 cm)
   55 keV: Si (40 cm) + S (20 cm)
   144 keV: Si (40 cm) + Ti (20 cm)
   These filters have been designed taking into account the MCNP results.

Six modules were irradiated, each at a pressure of either 1 or 2 atm and different filter; a seventh detector was irradiated, but with a polyethylene block inserted in the filter line in order to provide a measure of the scattering backgrounds. Measurements were performed at full (1 MW) reactor power, with a temperature ramping of the SDDs between –1° and 15° C at ~1.1° per hour.

Preliminary results for 1 atm are shown in Fig. 2. The structure at low temperature is clearly more consistent with theoretical expectations than the previous CERN $^{252}$Cf study. The predicted spectral kink in the Fe beam at 4.6 keV is clearly in evidence. The results are being analysed both following codes developed by Collar, and an MCNP simulation.

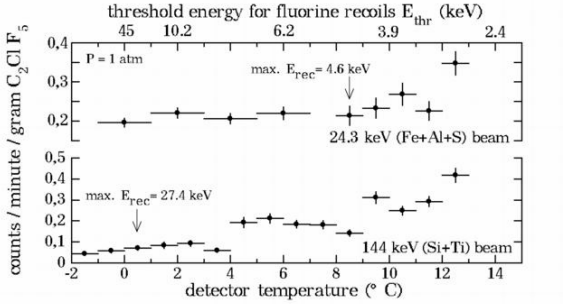


**Fig. 2** Preliminary results of quasi-monoenergetic neutron irradiations at RPI

## Published, accepted or in press work

1. $^{109}$Cd Irradiation of a Metastable Sn Foil: TA Girard, P. Valko, M.R. Gomes, C. Oliveira, G. Jung and V. Jeudy, in Proc. Particle Detection & Metastable Superconductivity, ed. TA Girard, M.R. Gomes, P. Valko (in press).
2. SIMPLE: Present Status and Improvements JI Collar, D. Limagne, T. Morlat, J. Puibasset, G. Waysand, HS Miley, TA Girard, F. Giuliani, C. Oliveira, J.G. Marques and F. d'Errico, in Proc. XIIIeme Rencontres de Blois: Frontieres of the Universe (in press).

[1] Centro de Física Nuclear, Universidade de Lisboa, 1649-003 Lisboa, Portugal
[2] Portuguese Research Reactor, ITN
[3] GPS-URA 85 CNRS, Universités Paris 7/6 25251 Paris, France.

# Electrical Discharges on Environment and Material Processing Applications

N.R. Pinhão, M.S. Silva[1], M. Pinheiro[2], K. Kutasi[3], A.C. Gonçalves[4], L. Redondo[5]

## Objectives

The main objective of this project is the use of cold plasmas in applied fields. As research and development in this domain is heavily based on numerical modelling as well as on experimental work, one goal has been to establish a solid experience on numerical modelling. The accurate description of the electron kinetics, through the solution of the Boltzmann equation, complements the chemical kinetics library developed last year and allows the development of self-consistent discharge models.

At the same time experimental work has been started on an applied project and will continue next year.

## Results

*Numerical modelling*

- A new fast code for the solution of electron Boltzmann equation in cold plasmas has been completed. This code is based on the two-term approximation and can be used to study both DC and RF discharges. Among others, it includes superelastic collisions, electron-electron collisions and takes into account non-conservative reactions.
- The application of a genetic algorithm to cross section fitting under simplifying assumptions has been successful allowing a significative reduction on time and improved accuracy in comparison to the manual fitting procedure.
- The simulation of a glow discharge in $N_2$ with a carbon cathode to the production of CN and $CN_x$ thin films has progressed.

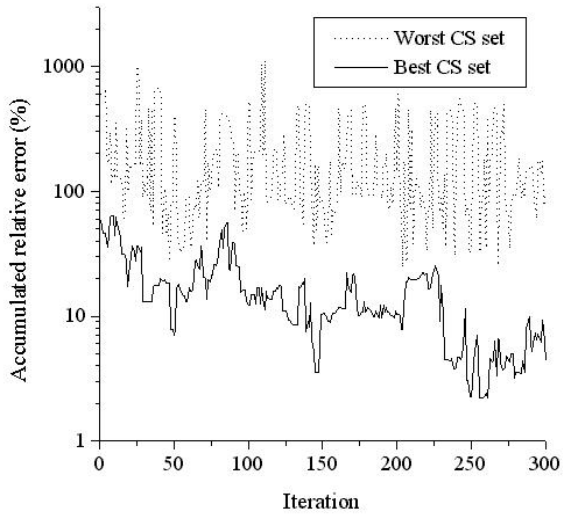


**Fig.** Application of a genetic algorithm: Total relative error for the best and the worst cross section set in each population for 5 control parameters.

*Experimental work*

- A laboratory dielectric barrier discharge system for gas cleaning has been projected and is under construction. This task is included in the project "Treatment and valorisation of plastic waste" A mass spectrometric gas analysis system for atmospheric pressure sampling has been projected and is been assembled. This system will allow the detection of concentrations on the 10 ppm scale.
- A Langmuir electrostatic probe has been built and preliminary measurements have been made on a low-pressure discharge.

## Published work (or accepted)

1. N. R. PINHÃO, PLASMAKIN: A chemical kinetics library for plasma physics modelling, *Computer Physics Communications* **135** (2001) 105-131.
2. N. R. PINHÃO, *PLASMAKIN Manual*, CPC International Program Library, catalogue Id: ADNH
3. L.M. REDONDO, N. PINHÃO, E. MARGATO, J. FERNANDO SILVA, Progress on high-voltage pulse generators, using low voltage semiconductors (<1kV), designed for Plasma Immersion Ion Implantation (PIII), *Surface and Coatings Technology* (in press)

[1] Last year student, Mathematics degree, FCT/UNL
[2] Centro de Física dos Plasmas, IST
[3] SZFKI; Budapest, Hungary
[4] Fellowship, Sociedade Ponto Verde
[5] Secção de Electrónica Industrial,/DEEA, ISEL

# Technical Assistance in the Field of Engineering Applications of Radiation and Radioisotopes

J. B. Manteigas, J. Neves, C. Cruz, F.G. Carvalho, J. Salgado

## Objectives

The main objectives are the development of equipment for internal groups, fabrication of equipment for specific applications and assistance to industrial companies and scientific institutions.

## Results

A summary of the more relevant work carried out is:

1-Design and construction of:

- A multifunction PCB with NIM bus for the Two-Axis Diffractometer, DIDE;
- A new DNG-P, Portable Gamma Level Indicator for controlling the level of bottled liquefied gases and for localisation of materials in pipelines;
- A new series of Personal Dosimeter RADX100 (SMD technology) – test and calibration of 100 units have been carried out;

2-Electronic assistance for nuclear instrumentation to DPSR, Physics Sector, UTR and Reactor;

3-Technical and scientific assistance for nuclear instrumentation to Solvay Portugal and Instituto Geológico e Mineiro.

A summary of the more relevant services and equipment rendered in 2001 is presented below.

### Summary of the more relevant services rendered in 2001

| Activity | Quantity | Client | Price* |
|---|---|---|---|
| Supply of Personal Dosimeters RAD X 100 | 2 | ITN/Reactor – DEA | 150 |
| | 1 | H. Sta. Maria/Porto | 75 |
| Supply of plating electrode disks | 250 | ITN/DPRSN | 50 |
| | 136 | ITN/DPRSN | 27 |
| Laboratory equipment for the determination of | 1 | Hellenic Labware, SA (GR) | 247 |
| radioactive element traces by electrodeposition | 1 | Neurotech, Inc. (USA) | 260 |
| | 1 | Florida State Univ. (USA) | 225 |
| Supply of gamma level detector units | 3 | CIMPOR/Souselas | 428 |
| Supply of Geiger-Muller Detectors | 4 | PORTUCEL/Tejo | 206 |
| Supply of portable gamma level indicator (DNG-P) | 1 | PREVINAVE | 600 |
| Measuring and control of source activities | 1 | CAIMA | 80 |
| | 1 | Siderurgia Nacional, S.A: | 30 |
| Supply of source containers | 1 | PORTUCEL/Tejo | 250 |
| *Technical assistance* | | | |
| - Troxler neutron gauge | 2 | ISA | 50 |
| - PVC modules | 6 | ISA | 64 |
| - Humicoque | 1 | Solvay Portugal | 30 |
| - Source container | 1 | PORTUCEL/V. Castelo | 370 |
| - Inactivation of radioactive sources | 1 | Siderurgia Nacional, S.A | 265 |
| - RAD X 100 | 2 | RITAGRA | 15 |
| - Boltzkin Software Library | 1 | Chemical Research Center (Hungary) | 230 |
| | | **Total Amount ($10^3$ PTE)** | **3 652** |

* Prices do not include TAX (IVA 17%)

# Radiation Technologies: Processes and Products

*Maria Luísa Botelho*

The **Radiation Technologies: Processes and Products** activities focus on the development and demonstration of radiation processing applications. Nowadays these activities are closely related to the gamma radiation facility (UTR), whose main applications are the sterilization of medical devices and pharmaceuticals and the decontamination of other products.

The group, therefore, supervises the sterilization and decontamination procedures being carried out at the UTR. This work is carried out in collaboration with **Food, Medical Devices** and **Pharmaceutical** Industries. The group also develops work with National and International normalization, standardization and certification bodies (IPQ, CEN and ISO). UTR upgrade and maintenance is performed in compliance with ISO 9002 in the perspective of obtaining accreditation.

In order to develop the procedures at UTR, based on the Quality System, studies of dose distribution and determination of Dmin and Dmax are performed taking into account the Safety Assurance Level and the safety of the product.
The Group develops work studying the impact of radiation on viable (e.g.: micro-organisms) and nonviable (e.g.: synthetic and natural polymers) materials.

The main purpose of the **microbiological** work **(viable material**s) is to develop and implement validation technologies for procedures of inactivation of micro-organisms, mainly by radiation (e.g.: $\gamma$ and $\varepsilon$ beam). These technologies are based on microbiological studies on the bioburden in/on the products, and aim to improve quality in this field.
Thus, hazard analysis and the control of critical points in the production lines of the studied products are part of the validation studies, carried out for the **Pharmaceutical** and **Food** Industries. Environmental control in surgical operation theatres at hospitals is also carried out.
In order to improve our understanding of the Radiation Procedures the action mechanisms of lethal agents in microorganisms are studied.

The **Syntetic** and **Natural Polymers** studies aim to use radiation both for developing new materials and for improving the quality of existing materials. These studies also aim to improve understanding about the interactions between radiation and these materials in order to avoid damaging the final product. Copolymerisation, reticulation and other effects induced by radiation are tested (carried out and characterised) to improve the properties and biocompatibility of new materials to be used for biomedical and environmental applications.

The Group collaborates with the University of Lisbon (Sciences Faculty/Plant Biology Department/ Molecular Biology and Genetics Group) and the University of Coimbra (Chemical Engineering Department).

A new field is presently being developed to apply **ionising radiation techniques** to **Wastewater treatment.** This technology could contribute to the correction and control of contamination by anthropogenically produced pollutants of water resources.

# Radiation Technologies: Processes and Products

## Research Team

### Researchers

- M.L. BOTELHO, Auxiliary Researcher (Group Leader)
- L.M. FERREIRA, Research Assistant (Responsible for the UTR)

### Collaborators

- M. H. GIL, Department of Chemical Engineering, Faculty of Science and Technology, University of Coimbra.
- R. TENREIRO, Departement of Vegetal Biology, Faculty of Science, Univerty of Lisbon.

### Students

- S. CABO VERDE, FCT PhD student of UL
- M. HELENA CASIMIRO, FCT PhD student of UC
- R. PATRÍCIA RIBEIRO, ITN Grant
- R. CORDEIRO, training student
- R. ZEINOU, IAEA fellowship, January to May 2001

### Technicians

- H.M. MARCOS
- P. PEREIRA
- V. DAMAS
- J. VENÂNCIO
- A. P. MARQUES (ITN Grant)

## Funding

| | $\times 10^3$ PTE |
|---|---|
| Research Projects: | 132 |
| UTR services: | 5942 |
| Expert services: | 350 |
| IAEA training fellowship: | 610 |
| **Total:** | **7034** |

## Publications

| | |
|---|---|
| Journals: | 1 and 2 in press |
| Internal Reports: | 3 |
| Proceedings: | 5 |
| Conf. Communications: | 2 |

# Products Sterilization

*M.L. Botelho[1], S. Cabo Verde[1], R. Ribeiro[1], A.M. Baptista[2], F. Russo[2], F. Águas[2], M. F. A. Proença[3] and M. Reto[3] and N.F. Teixeira[2]*

## Objectives

The aim of this field is to study the relevant parameters to guarantee the Safety Assurance Level SAL $10^{-6}$) sterilization of products at the UTR facility. Two kinds of pharmaceutical products were studied: TERRICIL ophthalmic ointment and medicinal "sea water".

## Results

The TERRICIL project was based on Hazard Analysis Control Critical Points (HACCP), carried out on the production line, and the subsequent determination of limits of initial contamination values for the alert or rejection of batch products. The determination of Dose sterilization (Dmin) of 17 kGy is based on real values. After the correction of critical points "Alert values" were determined and Dmin of 13 kGy will be enough to attain SAL of $10^{-6}$ (see **Fig 1**) [1].

Stability studies and the impact of gamma radiation on the activity of the drug (oxitetracycline) were also performed, in order to determine Dmax (see **Fig.: 2**) [2].

The "sea water" pharmaceutical product was studied in order to establish the impact of dose rate on the inactivation of natural contaminant microorganisms. Results using the turbidimetric technique (see **Fig.:3**) and $D_{10}$ parameter show that Dose Rate influences the inactivation rate of microbes (Lower dose rate →higher radioresistance) [3].

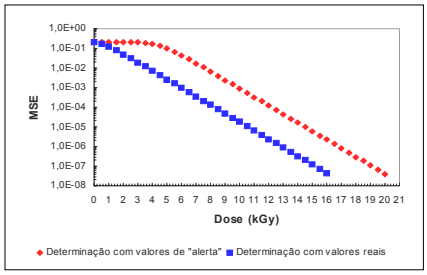


**Fig1.** Dmin *vs* SAL to alert values and real values.

**Fig 2.** Inhibition of oxitetracycline microorganisms before and after irradiation.

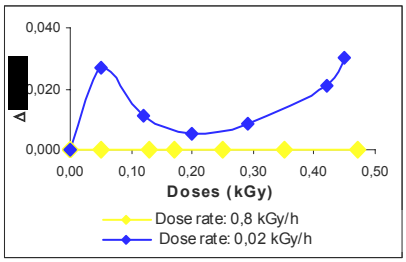


**Fig 3.** Turbidimetric results on "sea water" showing growth at lower dose rate local (0,02 kGy/h).

## Published, accepted or in press work

1. M.L. Botelho, A.M. Baptista S. Cabo Verde, F. Àguas, R. Ribeiro, and F. Russo, Estudo da Viabilidade de Esterilização por Radiação γ da Pomada Oftálmica TERRICIL-Selecção de Dose de Esterilização, presented in *2ª Jornadas da Sociedade Portuguesa de Ciências Farmacêuticas,* CCB Lisboa, Portugal, March 2001.
2. M.F.A. Proença, M. Reto, M.L. Botelho, O efeito da Radiação γ na Actividade da Oxitetraciclina, presented in *2ª Jornadas da Sociedade Portuguesa de Ciências Farmacêuticas,* CCB Lisboa, Portugal, March 2001.
3. R. Ribeiro, S. Cabo Verde, M.L.Botelho, Estudo da Cinética de Crescimento e Inactivação à Radiação γ de uma Estirpe Contaminante de um Produto Farmacêutico, presented in *2ª Jornadas da Sociedade Portuguesa de Ciências Farmacêuticas,* CCB Lisboa, Portugal, March 2001.

[1] Group of Radiation Technologies: Process and Products/ITN
[2] OFTALDER, SARL
[3] Dept. Tecn. Ind. Alimentares/ INETI

# Study of Thermal Properties *vs.* Gamma Irradiation of Disposable Surgical Clothing

L.M. Ferreira, M.H. Casimiro, C. Oliveira[1], M.E. Cabeço Silva[2], M.J. Marques Abreu[2] and A. Coelho[3]

## Objectives

This work is part of a study on the influence of ionising radiation (gamma and e-beam radiation) on the properties and performances of disposable materials for hospital protection.

In this case, our goals are to relate the results from thermal analysis techniques (DSC and TGA) with the mechanical properties observed in three materials (nonwoven, laminate and plyethylene textiles), after irradiation at several doses in a range from 0 to 200 kGy with gamma radiation from a Cobalt-60 source.

## Results

Differential Scanning Calorimetry (DSC) and Thermogravimetry (TGA) techinques show that nonwoven and laminate textiles maintain good molecular cohesion and do not show high levels of degradation for gamma radiation dose values lower than 100 kGy in nonwoven and 200 kGy in laminate materials.

The tensile strength and elongation decrease slowly for the nonwoven textile, and decrease faster for the laminate textile at 25 and 80 kGy absorbed doses.

It was found that for doses in that range, the increase in tensile strength observed in the laminate material is followed by an increase in the degradation temperature .

Concerning the nonwoven material the decrease in rupture strength is followed by a rapid decrease in temperature resistance, even at low doses.

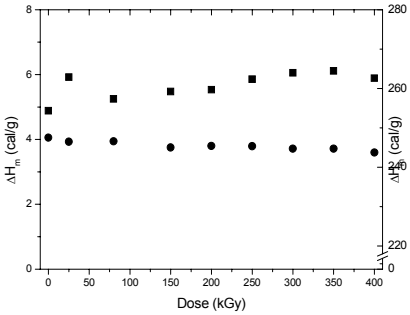


**Fig.** Variation of melting enthalpy ($\Delta H_m$- ■), and of melting temperature ($T_m$- •), with the absorbed gamma radiation dose, for nonwoven material.

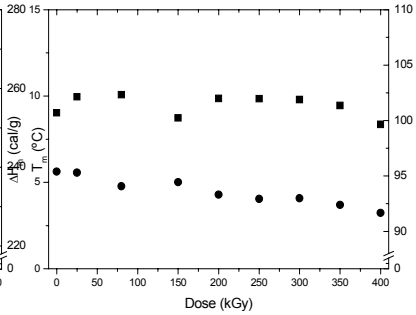


**Fig.** Variation of melting enthalpy ($\Delta H_m$- ■), and of melting temperature ($T_m$- •), with the absorbed gamma radiation dose, for laminate material.

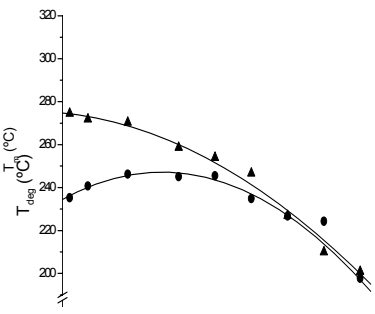


**Fig.** Variation of degradation temperature ($T_{deg}$), of nonwoven - ▲, and laminate- •, with the absorbed gamma radiation dose.

## Published, accepted or in press work

1. M.E. Cabeço Silva, C. Oliveira, , M.J. Marques Abreu, A. Coelho, I.F. Gonçalves and L.M. Ferreira. The Impact of Gamma and Electron Beam Irradiation Over the
2. Properties of Disposable Surgical Clothing, *Proceedings of the 12th International Meeting on Radiation Processing,* Avignon, France, March 2001.
3. L.M. Ferreira, M.H. Casimiro, C. Oliveira, M.E. Cabeço Silva, M.J. Marques Abreu and A. Coelho. Thermal Analysis Evaluation of Mechanical Properties Changes Promoted by Gamma Radiation in Surgical Polymeric Textiles, *Nuc. Inst. Methods B* (2001) In press.

[1] Nuclear Instruments and Methods group of ITN.
[2] Department of Textile Engineering, University of Minho, 4810 Guimarães, Portugal.
[3] FAPOMED, 4610-108 Felgueiras, Portugal.

# Food Irradiation

S. Cabo Verde, M. Luisa Botelho, R. Tenreiro[1] and J.Trigo[2]

## Objectives

Food irradiation is a promising technology for attaining safe products. In this field two projects are being developed: 1) **"Eggs"** and 2) "**Vegetables**".

1- **"Eggs"-** the aim of this project is to develop and validate methodologies for applying irradiation technology (radicidation/radiopasteurization) to eggs in order to get a safe product, mainly for *Salmonella* and *Campylobacter* spp..

2- "**Vegetables"** – the aim of this project is to evaluate the effectiveness of irradiation on microbial, colour, texture and sensorial parameters in some produce.

## Results

1-**"Eggs"** – Techniques and methods for the determination of bioburden in/on eggs were validated. Sampling of eggs was based on a random matrix in order to determine whether chicken age influenced egg bioburden. Results suggest that the egg bioburden is between $10^3$ to $10^6$ cfu/egg for the shell, and < 50 to $10^3$ cfu/egg for white and yolk [Fig 1]. Comparison of 50 eggs from 5 stages show that eggs from 65 week old chickens were the most contaminated both in shell and yolk/white egg. The more frequent contaminants were gram positive rods, catalase positive (47 %) and gram positive cocci, catalase positive (36 %) [Fig 2]. Until now *Salmonella* genus was not found. Although microorganisms of the *Kluyvera* genus and *E. coli* have been isolated on eggs and during HACCP studies (feed, drinking water, environment and faeces) the same genera were found on feed which point out the critical point. Further studies are carried out to confirm these results. Molecular methods will be applied to these bacteria to confirm genetic similarity. Studies for hygienization of feed by UV radiation are in progress [1].

2- "**Vegetables"** – preliminary experimental work has already been carried out in minimally processed vegetables. The methodology and work plan were discussed and presented at an IAEA coordenated meeting [2]

1- S. Cabo Verde, M. L. Botelho, R. Tenreiro "Avaliação da Carga Microbiana de Ovos de Galinhas de Diferentes Classes Etárias" presented in MICRO 2001, December Povoa do Varzim/Portugal.

2- M. J. Trigo*, E.S. Ferreira, M. M. Sapata, M. B. Sousa, T. Curado, L. Andrada, A. Ferreira, M. L. Botelho, and G. Veloso, Faculty First Research Co-ordination Meeting under the Coordinated Research Proramme on "Use of Irradiation to Ensure Hygienic Quality of Fresh, Pre-cut Fruits and Vegetables and other Minimally Processed Food of Plant Origin " presented in Rio de Janeiro, Brasil, November, 2001.

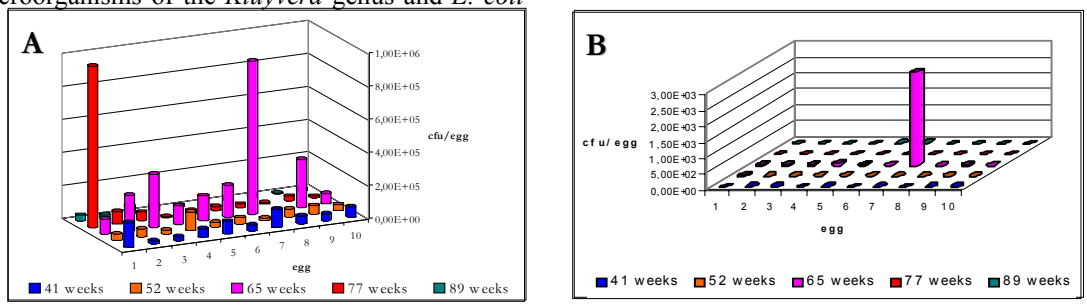


**Fig. 1:** Bioburden values (cfu/egg) for 50 eggs from 5 stages: (A) shell egg and (B) white and yolk.

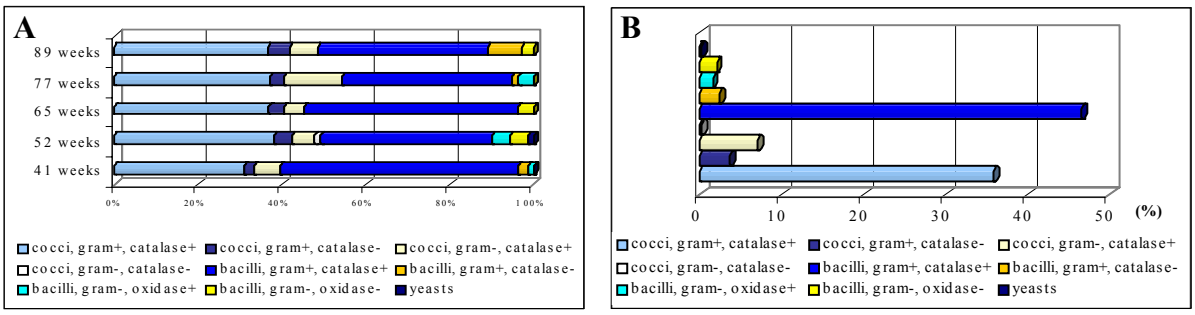


**Fig. 2:** Morphologic characterization of natural contaminants of 50 eggs: (A) in the 5 stages; (B) in the total sample.

[1] Department of vegetal biology, Faculty of Sciences, University of Lisbon.
[2] Instituto Nacional de Investigação Agrária

# Preparation and Characterisation of Chitosan Based Copolymers

M.H. Casimiro, M.L. Botelho and M.H. Gil[1]

## Objectives

Modification and characterisation of a natural biodegradable and biocompatible polymer (chitosan), in order to prepare copolymeric supports (hydrogels and films) to immobilise drugs in controlled drug delivery sistems.

## Results

Branches of poly(acrylic acid) were grafted onto chitosan using gamma radiation (from $^{60}$Co source), to initialize the polymerization. Two different systems of chitosan (solution of chitosan 3% in acetic acid 1%, and suspension of chitosan in water) were irradiated.

The influence of monomer concentration and the radiation absorbed dose on the yield of grafting were evaluated.

The characterisation of obtained copolymers was carried out by DSC, FTIR and water absortion.

The obtained results suggest that the yield of grafting increase with the absorbed dose and with the initial monomer concentration. And for small monomer concentrations, the copolymeric films swelling capacity seems to be independent from the studied dose values.

Study of graft copolymerisation of 2-hydroxyethyl methacrylate (HEMA) 10% onto chitosan (suspension in methanol) was also started.

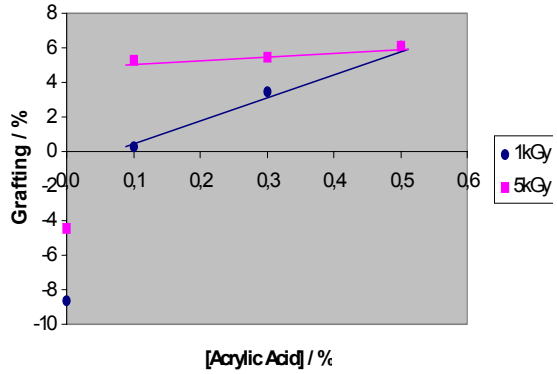


**Fig.** Influence of monomer concentration and radiation absorbed dose on the yield of grafting (powder samples).

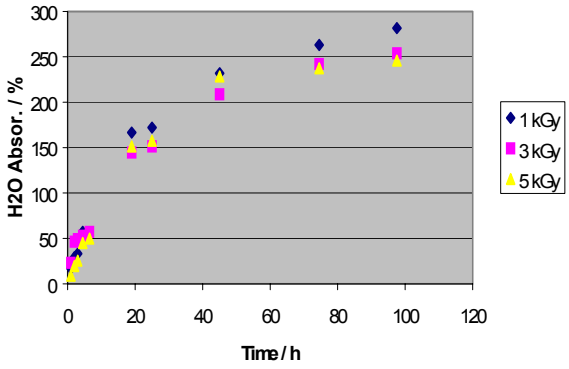


**Fig.** Water absortion of copolymeric films (Acrylic Acid = 0.1%).

## Published, accepted or in press work

1. M.H. Casimiro, M.H. Gil, Graft Copolymerisation of Acrlylic Acid onto Chitosan by Gamma Radiation, *Proceedings of 11th International Conference on Radiation Effects in Insulators*, Lisbon, Portugal, September 2001.

[1] Department of Chemical Engineering, Faculty of Science and Technology, University of Coimbra, Portugal

# Treatment of Wastewater by Gamma Radiation – A Case Study

M.L. Botelho, R. Ribeiro, R. Cordeiro, S. Cabo Verde, M.H. Casimiro, M.G. Ventura[1], C. Mustra[1], M.C. Freitas[1], M.H. Gil[2] and Ruba Zeinou[3]

## Objectives

Determine the impact of γ radiation on wastewater effluents. Two kind of studies were carried out

1) "Radiation Induced Degradation of Chloroorganic Pollutants in Waste Water - the objectives were to investigate the degradation of 4-ClBA under various experimental conditions using γ-rays, to identify the major products and to determine their yields (Gi – value).

2) This study was carried out on two kinds of municipal wastewater effluent samples, which are usually treated by UV irradiation. Microbiology studies were carried out to establish the response of micro-organisms to UV and γ radiation, as well as the influence of substrata and dose rate effects when natural contaminated effluent samples were irradiated at the gamma irradiation facility UTR. Preliminary studies were also developed using Fourier Transform Infrared Spectroscopy (FTIR).

## Results

1 - It has been established, that the substrate solution saturated with $N_2O$ (conversion of $e^-_{aq}$ into OH-radicals) are leading to higher degradation yields, compared to those saturated with air or pure oxygen. The highest degradation yield of 4-ClBA was, however, achieved (G=14,9) by irradiation under steady-state saturation with ozone.

At low dose rate (Dr =8,0 Gy/min) somewhat higher G-values for the 4-ClBA-degradation were observed in comparaison to those studied at higher dose rate. This effect can be explained by the fact, that at higher dose rate a larger portion of the primary products of water radiolysis are consumed by radical-radical reactions within the "spur".

2 - The UV treatment is seen to be more efficient to total coliforms than to total count. Furthermore the inactivation of micro-organisms by γ radiation at a higher dose rate (0.8 kGy/h) showed a decrease greater than $10^{-3}$ for total coliforms at 0.6 kGy. For total microorganisms there was a decrease greater than $10^{-5}$ at 5 kGy and the decrease was greater than $10^{-6}$ at 20 kGy.

The results of the total wastewater microorganisms' inactivation at lower dose rates could be seen to be somewhat less efficient than at higher dose rates. Determination of $D_{10}$ points to 90 % of the population irradiated at higher dose rates (0.8 kGy/h) was inactivated at 3.6 kGy, whereas at the lower dose rate (0.08 kGy/h) 90 % of inactivation was found at 16.4 kGy.

Simultaneous studies of bioburden on the non-irradiated sample throughout the duration of the experiment show that the microbiota of these samples show different characteristic microorganisms as the samples after irradiation at a low dose rate. The microorganisms of the irradiated sample reveal a homogeneous population of orange pigmented bacteria gram variable rods. This fact leads to the hypothesis that γ radiation could lead to the selection of more resistant species if the geometry and doses applied are not suitable/adequate.

The results of the inactivation effects of γ radiation on the liquid phase showed less survivors than the solid phase from the same sample after irradiation at the same doses.

The inactivation curves and $D_{values}$ show that the microorganisms demonstrate higher resistance on the waste water (WW) substrata than on the TSB substrata, probably due to components of WW that protect the microorganisms, for instance reducing agents, such as thiol groups. The bacteria present a higher $D_{value}$ when irradiated at Lower Dose Rate than Higher Dose Rate. One of the possible reasons is if conditions are conducive to microbial growth, the duration of irradiation is very important and furthermore, repair mechanisms of bacteria could respond more easily.

From the FTIR analysis one can suggest that there are no substantial alterations in the solid components; the spectra from the filtrate show that are some alterations (≈1420 cm-1) after irradiation which can be related to the presence of either thiol groups or nitrogen groups.

## Published, accepted or in press work

1. M. Luisa Botelho, N. A. San, C. M. Fldváry and Robert Kocjancic "Studies on Radiation Induced Degradation of chloroorganic Pollutants in Waste Water" IAEA Internal Report, under supervision of Prof. Nikolai Getoff, 3-21 September Vienna/Austria.

2. M.L. Botelho, Treatment of Wastewater by γ Radiation – A Case Study, presented at IAEA/RER 008, Istanbul/Turkey, IAEA/Internal Report.

[1] Atmospheric Elemental Dispersion.
[2] Department of Chemical Engineering, Faculty of Science and Technology, University of Coimbra.
[3] IAEA Fellowship

# *Radiation Technology Unit* - Irradiation Services

### *Industry*

| Client | Type of service |
|---|---|
| Jaba Farmacêutica, S.A. | Sterilisation of pharmaceutical devices |
| Oftalder - Prod. Farmacêuticos, S.A. | Sterilisation of ophtalmical dressings |
| Lecifarma, Lda | Decontamination of aromatic herbs |
| Vidrolab, Lda / Labmais, Lda | Sterilisation of PET flasks |
| Hovione - Sociedade Química, S.A. | Sterilisation of laboratory packing devices |
| Sapec Agro, S.A. | Sterilisation of |
| Falcão Teles, Lda | Decontamination of pharmaceutical raw materials |
| Hikma Farmacêutica, Lda | Decontamination of pharmaceutical raw materials |
| Others | Sterilisation of medical devices and pharmaceuticals, decontamination of raw materials and cork stoppers, irradiation tests of new industrial polymers. |

### *Universities and Research Institutes*

| Client | Type of service |
|---|---|
| Laboratório Agrícola da Madeira - Biofábrica | Dosimetric assays. |

### *ITN*

| Client | Type of service |
|---|---|
| Radiosterilisation Group (Physics Dept.) | Microbiological validation of radiosterilisation and decontamination.<br>Kinetic studies of microorganisms.<br>Waste water treatments by gamma radiation. |
| Polymers Group (Physics Dept.) | Irradiation tests of natural and synthetic polymeric materials |
| UTR | Study of irradiation geometries |
| Nuclear Instruments and Methods - Computacional Physics (Physics Dept.) | Irradiation tests for Monte Carlo simulations |
| *Project STERITEX* (Nuclear Instruments and Methods / Polymers Group and UTR) (Physics Dept.) | Dosimetric assays and irradiation tests of disposable medical devices |
| *Project Microstructure Investigation of Organic-Inorganic Hybrid Materials* - (Condensed Matter Physics / Polymers Group and UTR) (Physics Dept.) | Irradiation tests for the preparation of hybrid solid materials |

## *Irradiation services distribution at UTR during 2001*