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Tracer techniques as a contribution for studying the hydrological behavior of a São Francisco River sub-basin - PART I

Tracer techniques as a contribution for studying the hydrological behavior of a São Francisco River sub-basin - PART II

Tracer techniques as a contribution for studying the hydrological behavior of a São Francisco River sub-basin - PART III

#### Environmental tritium as a tracer for the verification of groundwater flow models

Cota, S., Velásquez, L.N.M., Minardi, P., Bomtempo, V.L.

The mathematical modeling of the ground water flow is a powerful tool to help understanding the dynamics of the water in aquifers, estimating long-term impacts of the water exploitation, planning the proper management of water resources and predicting the fate of contamination plumes and the effectiveness of remediation techniques. Usually, the process of building and calibrating ground water models requires a large set of data, including physical characteristics of the aquifer, water balance to describe the water inputs and outputs, hydrodynamic parameters and distribution of the piezometric pressure head, among others. Since the calibration process not necessarily leads to a unique and widely applicable model, it is advisable to verify how well the calibrated model is able to predict a set of data other than the one used for building the model.

This paper aims to present the verification of the ground water flow model of the Bauru aquifer by comparing the transit times calculated by the model and the ones estimated for water samples from wells exploiting the Bauru aquifer in the area of the municipality of Araguari, situated at 569 km west of Belo Horizonte, the capital of Minas Gerais state, Brazil. Located at the north-northeastern border of the mesozoic sedimentary basin of Paraná River, between Paranaíba and Araguari rivers, Araguari is mainly supplied for public use and irrigation by the exploitation of the Bauru granular aquifer, and, in a minor scale, Serra Geral volcanic fractured aquifers. Both aquifers are unconfined and the recharge is mostly provided by precipitation. The ground water flow model was developed using the MODFLOW model and transit times were calculated by the MODPATH model based on the modeled data.

Transit times for water samples were estimated by applying the well-established tracer technique based on environmental tritium (3H) technique, which allows determining transit times of ground and surface young waters (less than 300 years old). Tritium concentration of 51 samples of water (both ground and surface water) collected in 2004-2005 were analyzed and compared with the ones obtained in the precipitation of selected GNIP – Global Network of Isotopes in Precipitation/IAEA - stations (Brasília, Porto Alegre and Kaitoke), taken as input data for the exponential model considered in this work. Fifteen wells were selected to be compared with the model results, based on the existence of reliable information about depth and length of the filters and the exploited aquifers.

Calculated and estimated transit times for a significant number of selected wells showed a good correlation, indicating that the model was well calibrated and represents the actual behavior of the aquifer. The technique has shown to be useful to identify localized problems in the model. Lack of data for obtaining a more consistent estimative of transit times from tritium concentration, uncertainties related to the model choice for transit time estimative and integrated value of tritium concentration in multi-filter wells are some of the difficulties related to the application of this technique.

## Use of <sup>24</sup>Na in the determination of the moisture carryover in Angra 1 nuclear power station steam generators

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Angra Nuclear Power Station, in Brazil, is constituted of Unit 1 (657 MWe, reactor in operation since 13/mar/1982), Unit 2 (1350 MWe, reactor in operation since 14/jul/2000) and Unit 3 (1350 MWe, in construction). As part of the commissioning tests of Unit 1 it was measured, together with Westinghouse, the moisture carryover in the two steam generators, with the use of the radioactive tracer sodium carbonate Na2CO3 injected in the feedwater and collected in the steam sample lines leaving the steam generators. A correlation between the measured activities in the feedwater line and in the steam lines lead to the determination of the steam moisture carryover. The sodium carbonate sources were produced in the CDTN (Nuclear Technology Development Center) of CNEN, in Belo Horizonte-MG. Such commissioning test, carried out two times, confirmed the final result of 0,172 % moisture carryover with "steam probe", a temporary steam sampling collector, and 0.222% with sampling through installed Plant systems. Both results were less than the assumed value of 0,25% in the scope of supply of the steam generators. Each source were 4,5 g, 1,0 Ci and 2,0 Ci, measured during the addition in the feedwater. They were transported inside specific shield (338 kg), by truck, with special logistic arrangements taking into account the 10 hours transit time and the half-life of the sources (15 hours), with time enough for their valid utilization in the tests. During the tests the ETG - Turbine-Generator Building of the Plant was considered radioactive controlled area. In the first test the tracer was injected at 21:20 hours of 20/06/85 and at 21:20 hours of 23/06/85 the ETG was released for access without necessity for radiological control. At the 10:00 hours of 26/06/85 the area in the proximity of the tanks utilized for injection of the sources was also released. In 25 years of operation of the Plant, inspections do not indicate the presence of erosion of the turbine blades that could have been caused by excess of humidity in the steam, demonstrating the effectiveness of the tests performed during commissioning of the Plant. Also, the daily calculations of the reactor thermal power, which depend on the moisture carryover values have being demonstrated coherence.

### Application of radiotracer methodology to study mixing and segregation in a rotary mixer in glass industry

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Radioactive tracers are very suitable to study the homogenization process in a industrial rotating mixer because they can be measure through the wall with disturbing the mixing process. Tracer studies have been undertaken using a Eirich DZ29 mixer with 5.0 Ton cylindrical rotating chamber (2.8 m diameter and 7rpm), two mixing rotating star, one clockwise (38 rpm) and one anticlockwise (31 rpm) and a drum type rotor (484 rpm) and to measure the quality of final process 140La was used to mark three different components of the mixing process (sand , Sodium Sulfate and Iron Oxide) and two NaI 2x2" scintillators detectors were used do register the tracer signal outside of the vessel. It was possible to identify a dead zone inside the chamber and segregation in the mixing 'process.

## Determination of residence and mixing times through a slurry bed waxes reactor

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In a slurry bed waxes reactor, synthesis gas is converted to liquid hydrocarbons. This is done through a process wherein the gases react in a slurry of catalyst powder suspended in molten wax. In this study, the mean residence time of the gas through the reactor and the homogeneous mixing times of the liquid and catalyst in the slurry bed are determined by means of gas and liquid radioactive tracers. The experimental setup, the choice of tracers and the method employed are explained. The results are also presented and discussed.

Keywords: Residence time, mixing

## Comparation of blood residues effects in body tissues considering dose estimation three lutetium models

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The lutetium biokinetic models for the estimative of the patient dose are described in the ICRP 30 and ICRP 78 reports. However, none of those models subtract the blood contained in the organ and that circulating in the organ neighborhood of the organ. Alternatively, the biokinetics data can be obtained by 'post-mortis' measurements of the radioactivity in the organs. However, this kind of sample is appropriated only for studies in animals; is not possible get rid of the problem by removing all the blood on the organs. The present work is based on the Lutetium model described in the ICRP 30 and the Cerium model described in the ICRP 78 and the free Lutetium model presented by Jiménez. The software ANACOMPTM and the MIRD protocol were used to describe the transport of the element from fluids to other tissues and its elimination constant rate. The compartmental analysis theory helps to elucidate the dose overestimation due to the blood residues in the organs studies. Hypothetical values of 0%, 5% and 10% of blood contained in the liver, bone and kidneys were chosen to generate the dose response curves. Afterwards, these curves are used in the dose calculation, by interpolation of the dose in the tissues applying the reference values described in the ICRP 89 for regional blood contents in the bone (4%), liver (10%) and kidney (2%). These results show that the three models present different results in absorbed doses. The ICRP 78 model (0,28 mGy/MBq) presents results 52% upper than ICRP 30 model (0,19 mGy/MBq)and 29 times upper than Jiménez model (0,01 mGy/Bq) to whole body doses, while the organs doses growing for the ICRP 78 model and decreases for the Jiménez model. If the amount of blood in each tissue is not subtracted, the absorbed dose will be overestimated by ICRP 30 model in 0.08% (27.87 ightarrow27.89mGy/MBq); 17.2% (0.36  $\rightarrow$  0.43mGy/MBq) and 22.6% (0.23  $\rightarrow$  0.29mGy/MBq) to the bone, liver and kidneys, respectively. By ICRP 78 model the doses will be overestimated only to the kidneys in 41,72% (13.71 19.43mGy/MBq) and will be underestimated in 9.99% (13.38  $\rightarrow$  12.04mGy/MBq) and 9.88% (8.30  $\rightarrow$  7.48mGy/MBq) to the bone and liver, respectively. By the Jiménez model all the doses will be overestimated in 272% (0,05  $\rightarrow$ 0,20mGy/MBq); 180% (0.31  $\rightarrow$  0.84mGy/MBq) and 25%(0.26  $\rightarrow$  0.32mGy/MBq) to the bone, liver and kidneys, respectively

KEYWORDS: biokinetic model, compartimental analysis, MIRD protocol, dose estimation, blood in organ, Lutetium.

## Guide to radiopharmaceuticals interactions with other drugs

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Too many adverse reactions related to radiopharmaceuticals take place every day in hospitals routine, but many are not reported or even sensed. Information concerning these kind of reactions is not abundant and nuclear medicine staff is usually overwhelmed by this information. As every healthcare intervention carries some risk of harm, clinical decision making needs to be supported by a systematic assessment of the balance of benefit to harm. A systematic review that considers only the favourable outcomes of an intervention, without also assessing the adverse effects, can mislead by introducing a bias favouring the intervention that in the case o radiopharmaceuticals

may to render a important factor related not even to the quality of the drug but even to the quality of the diagnosis. The results suggest a logical framework to make decisions in reviews that incorporate adverse reactions. Researchers undertaking a systematic review that incorporates adverse reactions must understand the rationale for the suggested methods and be able to implement them in their review. Beyond a world effort should be made to report as many cases of false positive and adverse reactions with radiopharmaceuticals as possible. Only if this is done a complete picture of false positive reactions with radiopharmaceuticals can be drawn.

## Comparative study of the catalytic powder behavior at two different risers of FCC plants

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Comparative studies of the flow behavior of the catalytic powder at the "RISER" at two FCC Plants were made, using four radioisotope methods; radiotracers, gamma scanning, tomography and correlation flow measurement. These techniques allowed determining the internal behavior of the catalyst, the dispersion of charge and water vapor feed inside the RISER.

The internal behavior can be see in six graphs, two for the radiotracers, two for the gamma scanning and two groups of tomography graphics obtained at the two different FCC Plants.

Time contact of the charge and catalyst at the RISER was measure with radiotracers, the variations of density a long the RISER's vertical axis (high) were obtained with gamma scanning and several horizontal tomographyc cuts were obtained at different highs of the RISER. With this information, obtained from the two different RISER Plant design (denominated FCCI and FCCII), it was possible to evaluate and compare the fluid behavior at the two RISERs, that sustain the following assertions:

1. Different residence time distribution for the vapor and catalyst phase at the Riser.

2. Deficient mix between the reaction vapors and the catalyst, a long the vertical axis and horizontal cuts, that are due to canalizations at the two RISERs, showing that there are volumes were the catalyst is too concentrated and others were it is diluted, these volumes were along the vertical axis and also manifested in the horizontal cuts.

3. Under the last observation, and the analysis of the horizontal cut made above the nozzle section were supposed that the nozzle section did not operate uniformly, for this reason was necessary to measure the flow rate of water vapor and of the charge at each independent nozzle, that injects the charge to the RISER, to satisfy this aim the method "correlation flow measurement" was employ, finding that the nozzle 6 at FCCII had a double flow than the mean average of the other five nozzles.

After adjusting the control flow valves, for the charge and water vapor, for each nozzle, it was possible to increase 17% the production of gasoline, at FCCI. Also, it was possible to find out hydrodynamic design differences between both designs.

The paper present all the graphs obtained and their interpretation. This work was done at the IMP for PEMEX in 1998 and 1999.

[1] Colaborate with the Instituto Mexicano del Petróleo (IMP), until 2003.

## Industrial radiotracer generators

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Radiotracers are excellent tools for measuring flow and trace leakages in industrial facilities. Among the advantages are high accuracy, low detection limits, on-line measurements without perturbing the industrial process and well-known detector technology. Tracers with long-lived radionuclide may show disadvantages like disposal of waste and extended awareness of radiation protection of workers. If short-lived radiotracers can be used, there is no waste generated and radiation exposure may be minimized.

The radionuclides must be produced in a nuclear reactor, a particle accelerator (e.g. a cyclotron) or generated by decay from other radionuclides in so-called radionuclide generators. The latter are based on nuclear genetic mother-daughter relationships. In most cases the availability of a nuclear reactor or a cyclotron is limited, and for production of short-lived radionuclides the handling and transportation times are too long for the radionuclides to survive in many cases. However, a radiotracer generator based on a radionuclide generator is more versatile and useful for on-site production of short-lived radiotracers even at remote locations.

In a radionuclide generator the mother nucleusof one chemical element decays into a daughter nucleus of another element. Thus, they may be separated chemically. In a radionuclide generator the daughter (in most cases) is separated from the mother by elution (extraction) leaving the remaining mother nuclei untouched. The isolated daughter radionuclide may be utilized either directly as a radiotracer or as chemically bound in a tracer molecule.

If the half-life, of the mother radionuclide is long compared to that of the corresponding daughter radionuclide, the daughter will "grow inn" until secular equilibrium between the activities of the two nuclides are reached. The daughter nuclide can be repeatedly eluted because mother radionuclei produces daughter radionuclei continuously. The necessary time between elutions is exclusively governed by the half-life of the daughter radionuclide and the required amount of radioactivity for a particular purpose. Generally, this delay time should be longer than one half-life of the daughter. The operative period of a radionuclide generator is practically restricted to 2-3 half-lives of the mother radionuclide due to radiolytic damage and other chemical processes, which may reduce the milking efficiency and/or induce too high breakthrough of the mother and thereby affect negatively the operation safety.

There is presently a lack of applicable radiotracer generators for various industrial applications, and IFE has established a research program for this purpose. This program will utilize the following nuclear genetic relationships: 68Ge®68Ga, 113Sn®113mIn, 137Cs®137mBa and 144Ce®144mPr. These are chosen due to suitable nuclear properties (half-lives and radiation energy). The present paper is a status report on attempts to generate suitable radiotracers for tracing water and oil in oilfield operations, rather than studying the radionuclide generator systems themselves. This includes chemical complexation reactions and study of the stability (thermal, chemical, microbial) of the radiolabelled complexes as well as their behavior towards adsorptive surfaces, under simulated industry conditions. Our main focus at present is application in phase separators in the oil industry including the so-called scrubbers. Radiotracers both for the oil (organic) and the water phase are needed. So far good results have been achieved with water tracers of 113mIn-chelate complexes, and there are high expectations for synthesizing hydrophobic 113mIn-labelled oil tracers.

## Boron quantification in wood: an application of radiotracers

Celso Vargas (1), Mario Conejo (2), Roger Moya (3) Joon Ha Jin (4)

Protecting wood from attacks of termites and other agents is of relevance for wood industry. This protection is made by the introduction in the wood of some chemicals with specific properties. One important research subject is the development of new methods to effectively cure wood in a long lasting way. The other is the proposals of new methods for quantification the amount of chemicals absorbed by hydrophilic pores and other dynamic processes that take place in a wood piece, so that we may assure the permeability of wood to a specific chemical. The application of low energy and short half-life radiotracers, such as technecium-99, is an important method to achieve this quantification. In this paper we report our results in several tests conducted in the Non-destructive

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Storage of carbon dioxide in deep saline aquifers is a very promising solution to reduce our greenhouse gas emissions. In this regard, the behavior of caprocks in contact with CO2, either in supercritical state or dissolved in the brine initially contained in the aquifer, is of primary importance. Numerous phenomena can lead to CO2 leakages and, in order to guarantee the safety of the storage, a good knowledge of the caprocks transport parameters is necessary. This work focuses on the behavior of unaltered samples; all the tests reported here have therefore been done without CO2. This is the first step of the characterization; wherea s the influence of CO2 and its reaction with caprocks will be treated in future work.

Samples come from the Charmottes field in the Paris basin. Four layers, located from 1900 m and 2000 m depth, were studied. They are very compact rocks mainly composed of carbonates, with porosity varying from 2% to 5%, measured using mercury porosimetry.

Permeability experiments have also been performed. As the pressure of the CO2 bubble might be quite high, migration of carbon dioxide by permeation is possible. In order to be as cl ose as possible to storage conditions, triaxial cells have been used and a confinement pressure up to 300 bars has been applied to the samples. Intrinsic permeabilities have been measured using both gas and liquid experiments. For gas tests, a helium pressure gradient has been applied to dried samples and the permeation flow rate was measured by mass spectrometry. The results were corrected for the Klinkenberg effect and the intrinsic permeabilities were very low (under 10 µDarcy). For liquid tests, a synthetic brine has been used in a saturated sample and the flow rate measured by weighing. Some discrepancies have been found between the two measurements, and can be explained by the difficulty to obtain a complete saturation of these very low porosity materials.

Another transfer mechanism through the caprock is the molecular diffusion of carbon dioxide after its dissolution in the site water. The water self-diffusion coefficient has thus been measured using tritiated water as tracer, since it has a diffusion coefficient close to dissolved CO2. Through-diffusion method has been used with tritiated water as tracer. The results vary from  $1.10-11 \text{ m}^2/\text{s}$  to  $6.10-12 \text{ m}^2/\text{s}$ . This variation is interpreted to be related to the high heterogeneity within samples of the same core.

The samples studied are very tight rocks with a low porosity and it made all the measurements difficult and timeconsuming. However, it provides good and reliable characterization of the unaltered rocks. Similar tests will be done in presence of carbon dioxide and under more realistic storage conditions (higher pressure and temperatu re). Comparison of petrophysical properties of a "healthy" sample and an altered one will give us precious data on the behavior of rock in presence of brine and carbon dioxide. Y. Le Moullec, O. Potier, C. Gentric, J.P. Leclerc

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Two most different methods have been used for several years to model the hydrodynamics of reactors. The first one, often called "systemic modelling", is based on direct interpretation of tracer experiments. It consists in describing the flow behaviour using a combination of properly interconnected elementary reactors (plug flow, perfect mixing reactor, dead volume.). It is a global approach which gives quite rapidly and with moderate efforts a first approximati on of the reactor behaviour and can be used to simulate chemical reactions. However it remains unsatisfactory when complex local phenomena are involved and has only limited predictability.

For the last fifteen years, a second approach, computational fluid dynamics (CFD), has been used to simulate the hydrodynamics of chemical reactors. Owing to numerous developments and improvements, this approach allows a satisfactory description of the hydrodynamics but it still remains difficult to handle when it is desired to simulate hydrodynamics, heat and mass transfer and chemical reactions all together.

One intermediate solution is to find a reasonable trade-of f between accurate description of local phenomena and limited calculations by using informations derived from tracer experiments on the one hand and CFD simulations on the other hand. This idea has been used several times by developing complex compartment models derived from tracer experiments for which both elementary reactors and parameters are justified or estimated using CFD simulations. However these models remain a description of the main global trends of the reactor but not of the local effects which are sometimes preponderant. The objective of this work is to present a method to developed compartmental model based on both tracer experiments and CFD simulations and taking into account local description of flow, transfer and chemical processes. In order to illustrate the proposed method, the aerated channel sludge reactor has been selected. The pollution removal by micro-organisms is an essential step in the biodegradable wastewater treatment plants. These biological reactions often take place in channel reactors with horizontal water flow where the gas is injected at the bottom ensuring the mixing of the liquid phase. It consists in a cross flow gas/liquid reactor with a very long length compared to its height and its width. Due to its shape and size, the efficiency of depollution reaction is significantly affected by hydrodynamics. Therefore, due to the complex coupling of hydrodynamics, heat and mass transfer and biological reactions, it is a perfect reactor to test the robustness of the proposed approach.

The mapping of the compartment modelling has been determined following three rules: flow should not change direction along a compartment boundary, physical scales should not deviate too far from an average value in each compartment and the compartmental model RTD should mat ch the experimental one. Flowrates connecting compartments have been determined from CFD mean flow field and turbulence characteristics. Oxygen transfer has also been determined from CFD simulation. The simulated biological phenomena have been compared with experimental data in terms of evolution of chemical oxygen demand (COD), suspended solid (SS), total nitrogen (TN) and oxygen concentration (SO) along the length of the reactor with a satisfactory agreement of both global and local concentrations. The proposed method can be extended to numerous complex reactors for which CFD approach still remains too complex and classical systemic model derived straightforward from tracer experiment cannot be extrapolated?

## Comparison of several models and/or software's for tracer test interpretation in a five spot laboratory set-up

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Tracer test interpretation in oil field is a difficult challenge due to the complexity of the natural media, the lack of information, the low and incomplete tracer recovery. It is first recommended to test the chosen models in the simplest possible configuration before interpreting the data issued from oil fields. Because of this an experimental five spot setup has been designed. It is a large rectangular vessel of 160 cm in length, 95 cm in width and 80 cm in depth. It has been filled manually with sand with a density of 1.8 and an average granulometry of 0.75 mm (d10=0.5 mm, d50=0.75 mm, d90=0.90 mm). The injector-producer distance is only 40 cm to avoid boundary effects with 5 wells of 40 mm diameter. The bed of sand is initially saturated with water. Four experiments have been conducted, three experiments with stagnant water inside the bed and flow-rates of 107 cm3/min (exp 1 and 2), and 184 cm3/min (exp 3) in the injector well. The fourth experiment has been conducted with a linear velocity of the water table of 2 meters/day and proved difficult to exploit. The data have been time and area normalized in order to prevent the effect of the non homogeneous radial concentration of inlet injection. If the main key parameters: breakthrough time, first moment and percentage of tracer recovery remain the same, the tracer concentration varied from one experiment to another one due to the non homogeneous radial injection. Four models have been tested:

Classical model for flow and transport in porous media using the Comsol and Castem finite element software packages Brigham model

Streamlines model using the Poro software package

The paper presents the whole set of simulations and comparisons with experimental data. The four models give a reasonable prediction of the experimental data obtained at laboratory scale but they present different advantages or drawbacks. The Brigham model is a very simple model but it gives access to global parameters directly interpretable by oil producers. CFD codes (i.e. Comsol and Castem) give both satisfactory agreement even if Comsol seems to be more accurate; however they request more calculation time which can become a real limitation for oil field tracer experiment simulation. Moreover they request a deep knowledge of the oil field structure. The Poro software based on streamlines model seems to be the best compromise. However, several model configurations may lead to the same tracer response which requires careful data interpretation procedure.

## <sup>63</sup>Ni(CN)<sup>4</sup><sub>2</sub>- and Ni(<sup>14</sup>CN)(CN)<sup>3</sup><sub>2</sub>-complexes as possible water tracers: Synthesis, purity assurance and analytical procedures

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Cyanide, CN-, is known to be a strongly bound ligand in many metal complexes and give the complexes high stability constants. Previous studies of the cobalthexa-cyanide complex, Co(CN)63-, led us to conclude that such cyanide complexes with high complex constants may be applicable water tracers but not in any environment,- there may be some serious limitations.

These studies were followed by similar investigations of other metal cyanide complexes like Ni(CN)42- with the possible radiolabels 63Ni and 14C, Fe(CN)63- with the possible radiolabels 55Fe, 59Fe and 14C, Ag(CN)2- with the possible radiolabels 110mAg and 14C and and Au(CN)4- with the possible radiolabels 195mAu and 14C. In addition, the two latter complexes may be used as non-radioactive tracers in combination with molecular enrichment techniques and sensitive detection techniques for Ag and Au, respectively.

The present paper deals exclusively with analytical issues of radiolabelled Ni(CN)42-. In development of analytical procedures for this complex, there are two main application areas:

The analysis of laboratory samples during the studies of the molecular (tracer) behaviour under simulated reservoir conditions, and

2. Analysis of field samples which require enrichment procedures before final quantitative detection.

For the first category we will report typical results from:

- Separation by electrophoresis of the radiolabelled complex in combination with a position-sensitive detection technique used on the electrophorogram, and of the non-radioactive complex in combination with sectioning of the chromatogram and neutron activation analysis of the sections.

- Separation by ion-pair chromatography

For the second category results will be reported from:

- Enrichment of the complex from produced water samples by separation on anion exchanger columns followed by elution and subsequent detection by liquid scintillation spectroscopy

#### Liquid and gas residence time distribution in a two-stage bioreactor with cell recycle

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The application of three phase reactors (solid, liquid and gas) is well established in a wide variety of chemical processing operations such as, production of fine chemical, coal wastewater treatment and fermentation processes. In our laboratory, high cell density cultures in order to perform high ethanol productivity were investigated with an original two-stage continuous bioreactor with cell recycle (TSCB) [1, 2]. The first and the second stage were respectively dedicated to cell growth and ethanol production. The high cell density was obtained by an ultra filtration module coupled to the second reactor. The innovative part of this bioprocess consists of the use of a recycle loop from the second stage to the first one to improve cell viability and activity (figure 1). The performance of the reactor is, however, strongly influenced by the complex interaction between gas and liquids hydrodynamics as well as the solids (microorganism) activities and mixing pattern [4]. In this work, our purpose is to study, characterize and model the residence time distribution (RTD) of liquid and gas phases in agreement with identified operating conditions with biological activity.

Figure 1. Experimental set-up (continuous two-stage bioreactor with cell recycle) and instrumentation dedicated to gas and liquid RTD study.

Figure 2: Residence time distribution, E(t) and Internal age distribution, J(t) in TSBC for liquid phase (Operating condition  $n^{\circ}1$ ).

RTD experiments were conducted with TSCB without biological activity and following the methodology described by Thereska [5]. Operating conditions enable to investigate the impact of gas flow rate, as well as liquid feed, extraction, recirculation and agitation. The range of operating parameters was investigated according to process mode: batch and continuous with and without recycle.

The tracing methods were respectively a step of oxygen or azote concentration as gas tracer and a pulse (Dirac) of NaCl/NaOH concentration as liquid tracer. For liquid phase, conductimetric and pH-metric methods were used to quantify tracer concentration in different locations in the bioreactor. The injected tracer quantity was controlled. For gas phase, percentages of oxygen concentration at the outlets of reactors were measured with two gas analysers (figure 1). The experimental measurements included flow rates, temperature, conductivity, dissolved oxygen concentration, pH, relative pressure and mixing rates. All sensor signals were electrically conditioned and collected using an on-line data-acquisition system.

In a first step, data analysis and RTD formulation were realised. Normalised and reduced experimental y(t) an y(q) signals were deduced from experiments. The experimental residence time distributions, E(t) or cumulative, F(t) and the internal age distribution, J(T), were obtained (figure 2) and characterized by a set of moments (mean residence time) and centred moments of order j (reduced variance for j=1). A dimensionless representation of E(q) vs q enabled to demonstrate the single hydrodynamic behaviour of TSBR for gas phase.

Figure 3: Experimental and model E(t) in first and second stage with gas phase (Operating condition n°1).

In a second step, the reactor behaviour was described with a RTD analysis based on DTS Progepi v4.2 software [3]. A systemic analysis was performed to simulate the global behaviour of the bioreactor (figure 3). Two different models were assumed for gas (CSTR in series) and liquid (CSTR in series with exchange and plug flow reactor) phases. For each model, all constants were identified by fitting the experimental curves and discussed versus operating conditions. RTD formulation stands as an efficient tool to give new insights in hydrodynamic phenomena occurring in the two stage bioreactor with cell recycle. Further investigations will focus on modelling each constant versus significant experimental factors (aeration, mixing, flowrate) and comparing RTD to biological performances.

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#### Use of terrestrial epiphytic community to biomonitor of atmospheric pollution at steel valley region, Minas Gerais State

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This work was carried out for verification of the inorganic contaminated presence in atmospheric pollution through a biomonitor of metals in industrial area pollution of the Steel Valley region, Minas Gerais State, Brazil. The studied cities has been: Ipatinga; Fabriciano Colonel; Timóteo; Santana do Paraíso e Marliéria. Terrestrial epiphytic community samples has been used as biomonitor. The samples were collected in trees Oiti (Licania tomentosa) and Angico (Piptadenia rígida), very common in studied region. The samples were collected in 17 points and two weather stations: January (rainy) and June (dried) of 2007. The samples has been analyzed in the Radiochemistry laboratory at the Center of Nuclear Technology Development (CDTN) of the National Council of Nuclear Energy (CNEN) in the capital of Minas Gerais State, Belo Horizonte. The Neutron Activation Analysis was used to determine the elements Al, As, Au, Br, Ce, Co, Cr, Cu, Fe, K, La, Mg, Mn, Na, Sm, Sr, Ti, Th, Zn, U and V. The k0 parametric method was used to determine the elements Al, Au, Co, Cr, Cu, Fe, Hg, Mn, Mg, Cr, Zn, V and Th when compared with the values cited in the literature. The biomonitor used in this work, terrestrial epiphytic community, showed a excellent capacity for metals retention by atmospheric contamination.

Keywords - biomonitoring, terrestrial epiphytic community, metals, atmospheric pollution.

## Chemical and biological characterization of the lutetium-177 labeled bombesin derivative BBNp6

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Radionuclides coupled to receptor-specific peptides are currently under investigation in clinical trials involving different tumors. They specifically localize receptors overexpressed on the membrane and then internalize into cells. In designing radiometal-based peptides for cancer diagnostic and treatment, important factors to consider are the physical characteristics, cost and availability of the radionuclide and the radiolabeled molecule chemical and biological properties. 177Lu-labeled peptides are attractive in diagnostic and targeted therapy of small tumors or metastases owing to the excellent radiophysical properties (half-life 6.65 days, medium energy b- 497 KeV, g radiation of 208 KeV) and commercial availability of the radiometal. In the field of radiolabeled peptides, bombesin analogs have become focus of interest because their receptors, specially gastrin-releasing peptide receptor (GRP), have been shown to be massively overexpressed in several human tumors, including prostate, breast and small cell lung cancers. In this work we describe the radiolabeling with 177Lu and some properties of the novel bombesin analog BBNp6 - DOTA-X-BBN(6-14), where X is a spacer of six aminoacids – in order to predict its target to tumor cells in animal tumor models. BBNp6 was planned by our group and purchased from piChem. Preliminary studies were done to determine the best labeling conditions and instant thin layer chromatography (ITLC) and HPLC were applied to evaluate the radiochemical purity of the preparations. The stability of the radiolabeled peptide was assayed either after storing at 4°C or incubation in human plasma at 37°C. Chemical characterization of 177Lu-labeled BBNp6 was done by partition coefficient determination. Finally, pharmacokinetics studies were performed at different time intervals after i.v. injection of 177Lu-DOTA-X-BBN(6-14) in normal Balb-C mice and the pharmacokinetic parameters were determine by Biexp software.

BBNp6 was labeled with high yield (98.89 + 1.1 %) after reacting with 92.5 MBq of 177LuCl3 at  $90^{\circ}C$  for 30 minutes and this mixture kept stable for more than 96 hours at  $4^{\circ}C$  and 4 hours in human plasma. This peptide exhibits low lipophilicity, in according to its partition coefficient, and in vivo pharmacokinetic studies showed a bicompartimental distribution model with fast blood clearance (half-life of the fast fase 0.22 h). These results suggest that BBNp6 has ideal characteristics for a radiopharmaceutical and can be applied in preclinical studies.

Further studies are in development to evaluate BBNp6 target to human prostate carcinoma PC-3 cells in Nude mice. Modifications at its spacer will be also investigated to improve in vivo properties.

## Effect of the medicinal plant echinacea on the labeling of red blood cells with technetium-99m

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The use of plants for the treatment of different diseases is a spread custom in the popular medicine and has increased in the last decades all over the world, but in some cases the biological effects of these plants are not known. The drug therapy could affect the blood elements labeling with technetium-99m (99mTc). This process has several important applications in Medicine like cardiovascular system analysis and detection of hemorrhage sites. Echinacea purpurea (Echinacea) is the most widely used herbal medicine in the treatment for acute upper respiratory infection based in its immunological properties. The aim of this study was to evaluate the influence of the extract of Echinacea on the morphology and labeling of red blood cells (RBC) in an in vivo study. Samples of heparinized blood were withdrawn from Wistar rats treated with Echinacea (180mg/kg) during 60 days and from the control group. Blood smears were prepared and evaluated in an optical microscope. The radioactivity in RBC were determined in a well counter and the %ATI was calculated. The analysis of the results shows that the treatment with Echinacea during 60 days could decrease the levels of RBC and hemoglobin, decrease the fixation of 99mTc in the RBC and alter the shape of RBC in comparison with the control group. The effects founded were statistically significant (Mann-Whitney test, p<0.05) and they could be explained by the biological properties of this studied extract of Echinacea.

Key words: Echinacea, red blood cells, morphology, technetium-99m.

## Development of a method for labelling mud with <sup>99m</sup>Tc, to be used in hydrodynamic studies of fine sediment in suspension – Application in Brazil

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The 99mTc, broadly applied in Nuclear Medicine, was studied in laboratory with the objective of labelling mud, through the chemical reduction of the TcO4- eluted from Mo/Tc generators (BANDEIRA, 2004). The 99mTc has T1/2 = 6.02 h and y-ray energy = 140 keV, being suitable for studying the transport, in water environment (rivers, estuaries, bays and open coast), of fine sediment in suspension. The labelling is followed by injection and subsequent in situ detection, by means of scintillation detectors transported at suitable depths, by positioned boats, allowing the determination of advection, dispersion and sedimentation rate parameters. Formerly, this kind of labelling was performed mainly using 198Au, 46Sc and 51Cr.

This kind of study associated with the knowledge of the hydrodynamic agents, e.g.: river, tidal, wind and wave currents are powerful tools for the understanding and quantification of the sediment transport in suspension. It also gives response to human interventions, such as: dredging of reservoirs, access channels and harbours, and dumping of the dredged material, in the water environment. Besides that, it contributes for the optimisation of the dredging works, allowing the evaluation of dumping sites and also the physical environmental impacts of the dumping, being a valuable tool for calibrating mathematical models of sediment movement.

The application of this technique will be shown for the dredging of the accreted Pampulha reservoir in Belo Horizonte-MG, Brazil, and the dumping of the fine sediment in the watercourses downstream its dam, the natural way for the sediment in its absence, searching for a perennial solution for the problem. Field experiments, with simultaneous and instantaneous injections of sediment and water labelled, respectively, with 99mTc and Rhodamine WT, were performed, in dry season, to measure the hydrotransport capability in a stretch of 25 km, since Pampulha creek until Velhas River, inclusively. They allowed comparing the different hydrodynamic behaviours of the mud in suspension and the water transporting it. A recent mathematical model was applied and calibrated to the data obtained and, through convolution the sediment dumping using a hydraulic dredging system was simulated, calculating also the physical environmental impacts: increase of sediment concentration and the possibility of deposition. Through the measurement of physical-chemical parameters of the water, the possibility of desorption of the metals adsorbed in the sediment to be dredged, was evaluated. It was concluded that there is no environmental impediment for the dumping of the dredged fine sediment in the watercourses downstream.

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In April 2006, under the framework of IAEA TC Project VEN/8/019: "Management of Sediments throughout the Navigation Channel of the Orinoco River" radiotracer studies were performed in Orinoco River, Venezuela, in the stretch Guarguapo-Barrancas-Ya in order to evaluate bottom and suspended sediment transport. The broad objective of the former study was related with the choice of a dumping site for the material dredged from the ship channels. The specific objective was to study the following characteristics of the behaviour of fine sediment in suspension: advection velocity, dispersion coefficient, sedimentation rate and dilution, taking into account that the fine sediment is the main carrier of heavy metals and other pollutants in the water environment. The Orinoco River basin region is presently undergoing a fast industrial development with many industries being installed in the river margins with outfalls discharging into the watercourse.

In relation to the suspended sediment study, two sub-superficial injections of mud labelled with 99mTc were performed. The initial activities used during the injections were respectively, 2.1 and 1.6 Ci. The detection was performed by a boat with two scintillation detectors placed at 1.5 m and 0.5 m below the water surface.

The advection of the suspended particles was determined from measurements of the displacement of the puffs of radioactively tagged particles. Thus, the distance from the injection point to the position of the maximum count rate of each crossing of the cloud, was plotted as a function of the time from the injection. The advection velocity was obtained by the inclination of the straight line adjusted to the points of this graph.

Each crossing of the radioactive cloud provides an approximately Gaussian distribution of the count rates. The dispersion coefficient is related with the rate of variance growth as the cloud expands. This increase could be expressed by a general law with the form of Var=Atm where A and m are adjustable constants obtained by least squares fit. Taking into account the regression coefficient values of the present study, one could assert that this function is suitable to represent the growth of the variance with time.

The sedimentation rate is determined from the decrease of the maximum count rate with time. In this way, the maximum count rates obtained for each cloud crossing were plotted, in a logarithmic scale as a function of time, and the envelops to the peaks were drawn. These envelops approximately correspond to the true variation of the peaks with time since the crossings only occasionally hit the maximum concentration of the tracer.

The minimum dilution attained at a given point of the water body is the relation between the initial concentration of the discharged solids and the concentration at this point. Hence this dilution can be calculated from the maximum count rate values determined from the cloud crossings.

The results obtained for the behaviour of the natural sediment in suspension, at the end of the low water season of the Orinoco River (April), could be used for preliminary designs of outfalls for industrial effluents which will discharge particulate material with a density similar to the fine sediment or for pollutant material that can be adsorbed bin the fine sediment.

#### Radiotracer application as a diagnostic tool in industries

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Citing experience in the South African industrial environment, a number of case studies in the areas of a liquid, gas and solid tracer's applications are presented. These are: The transit times of pollution in a river by means of radioactive liquid tracer technology, total residence time evaluation of slagment and metal in a sub-merged manganese-chromium furnace and the determination of the performance of waxes reactor in the petrochemical industry.

Keywords: Residence-time, irrigation, optimum homogeneous mixing, waxes chemical reactor, slagment, submerge furnace

# Radiolabelled Co(Cn)<sub>6</sub><sup>3</sup>- complexes, - possibilities and limitations for tracing water flow in oilfield operations

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There are not many records in the open literature about the use of metal cyanide complexes as tracers in industrial and geospheric tracing. However, it is well known that cobalthexacyanide has been used as a water tracer in the oil industry for many years. The co(cn)63--molecule is particularly interesting because of the fact that from this single molecule one may produce six analytically distinguishable species which may operate as individual tracers. Five of them may be radiolabelled with the radionuclides 56co, 57co, 58co, 60co and 14c, respectively. The sixth tracer is simply the non-radioactive molecule itself which has a high analytical sensitivity when enrichment techniques for the molecule is combined with elemental analytical techniques like, for instance, thermal neutron activation analysis (thnaa) or inductively coupled plasma in combination with mass spectrometry (icp-ms) for detection of the co-atom. The performance of the co(cn)63--molecule in oilfield applications has, however, been variable. In some applications there has been no difficulties detected while in others one has experienced non-performance and even heavy

there has been no difficulties detected while in others one has experienced non-performance and even heavy contamination of the injection equipment. These apparently contradictory results are obviously connected to the stability of the complex in the varying chemical environments and led to a more detailed investigation of the stability of the co(cn)63--complex as a function of appropriate parameters.

In the present paper we will report results from experimental investigations on synthesis of the radiolabelled complex, radiolytic stability, stability against sorption to various relevant materials (steels and reservoir rock surfaces) as a function of temperature and its analytical detection in samples of produced waters in ultra-low concentrations. In all these investigations (except from the final step on instrumental detection of radiation) we have used 60co(cn)63- as a laboratory tracer and representative for the complex.

## <sup>195m</sup>Pt- labeled cisplatin as a possible tool for glioblastoma treatment

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The antitumor activity of cis-dichlorodiamineplatinum(II) (CDDP, cisplatin) was discovered by Rosenberg et al [1]. However the effectiveness of cisplatin against recurrent tumours is lower than that against primary tumours [2], probably because of the presence of a population of cisplatin resistant cells as occur in gliomas. Gliomas are the most common and most deadly primary tumours found in the brain and carry a particularly poor prognosis. Because of its location beyond the reach of local control when it is first detected, these tumours have frustrated almost every attempt for successful therapy. By introducing radionuclides into the cells during the treatment with cisplatin, internal radiation and chemotherapy are possible at a low rate of toxicity. The proposal of this work was to investigate the antitumoral effect of neutron activated cisplatin at the TRIGA MARK-I IPR-RI and verify if the low-dose internal radio-chemotherapy produces additive effects on malignant glioblastoma cells.

Neutron activation was done on cisplatin as described by Leal et al.[3]. Briefly, cisplatin was irradiated at 100kW during 8 hours into polyethylene flasks carried out on a TRIGA MARK-I IPR-R1 nuclear reactor at the Centro de Desenvolvimento da Tecnologia Nuclear - Comissão Nacional de Energia Nuclear (CDTN-CNEN), Brazil. Chemical stability after neutron activation was evaluated on gel filtration chromatography. Glioblastoma cells were incubated with different concentrations of non-radioactive or radioactive cisplatin (195mPt-cisplatin). Cytotoxicity and apoptosis were evaluated after 48 h treatment and anti proliferative effect was evaluated by clonogenic assay.

The final specific activity for 195mPt-cisplatin after neutron activation was approximately 87kBq mg-1. Treatment with neutron activated cisplatin (IC50= 1.8 x10 -6M) proved to be more potent than native cisplatin (IC50= 4.9 x10 -6M) and irradiation alone (IC50 ~ 6 Gy).

The results obtained in the present work indicate that 195mPt-cisplatin kept its chemical stability upon neutron activation and was a very potent radiosensitizer evoking a supra additive effect. Treatment with internal radiochemotherapy based on neutron activated cisplatin became possible a significant reduction of the cisplatin concentration required for effective inhibition of glioblastoma growth. Therefore production of radioactive cisplatin based on neutron activation may constitute a good strategy for the preparation of novel therapy for malignant glioblastoma.

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## New method for preparing a <sup>35</sup>S in the form as a sulfite to be used as Tracer in medical and physiological research

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Labeled sulfite is used in preparing 35S labeled medical and physiological chemical compounds which may provide information on effectiveness and distribution of these compounds [1]. As sulfite, the labeled 35S is stabilised as a sodium salt which is chlorinated to form sulfanyl chlorides. This is further reacted with amines to form sulfonamides [2]. The many uses of sulfonamides include acting as competitive inhibitors of the enzyme dihydropteroate synthetase, preventing folate synthesis which is required for the bacterial cell to synthesize nucleic acids. Another important aspect of a (methyl) sulfonamide is that it led to a significant increase in the activity of spironoidanyl piperidine, a growth hormone secretagogues receptor [3].

35S labeled Methyl sulfonic acid is used as the precursor for 35S labeled methyl sulfanyl chloride and this acid is normally prepared from two main techniques;

The reduction of Ba35SO4 to 35SO2 with red phosphororus.- this chemical reaction is violent and difficult to control. [4]

Pyrolytic decomposition of 35S labeled copper sulfate – better control is achieved but requires high thermal heat and long duration.

In this method the pyrolytic decomposition of Cu35SO4 was achieved with an electric arc and the surface of a copper electrode was use to form the Copper (II) 35Sulfate pyrolised to 35SO2.

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> Tracers for wormhole characterization and gel treatment design and evaluation, in oil secondary recovery

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Heavy oil production in unconsolidated sands constitutes a complex task because of the oil - sand combined production. During the primary exploitation, the voids generated by the sand extraction contribute to increase the drainage volume of the wells and so the oil production. In contrast, during the secondary recovery of oil, when water is injected for pushing the remaining oil, the ends of the voids located far from the production wells (known as wormholes due to their resemblance) may be reached by the water injection. In consequence, the injected water will be being channeled between the injector and the producer, without pushing the oil banks. The usual corrective treatment consists of the wormhole plugging employing gels. During the gel treatment design, precise values of permeability and volume of the wormhole are necessary for optimizing the gel viscosity and quantity. For this reason, a pre-gel tracer test was carried out. Also after the gel injection a second tracer test was conducted for evaluating the gel corrective action. In both cases, the tracer records have showed extremely short breakthrough times and excessive mixing. Several models were analyzed for reproducing the tracer behavior and parameters evaluation. The best behavior was shown by a simple perfect mixing model, with mixers in series and diffusive exchange with the walls.

## Scintillating setup for high accessibility detection of low activity ionizing radiations

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We have developed a new family of miniaturized setups for the detection of ionizing radiation (alpha, beta, gamma, X). The device consists in a plurality of high accessibility scintillation probes. Each probe is made of a scintillating crystal optically coupled to several optical fibers. The outputs of these fibers are detected in coincidence by photomultipliers: a light pulse emitted by the crystal will be validated if photons are detected by each photomultiplier simultaneously. This selection allows considerable reduction of the effect of the "dark counts" of the photodetectors and thereby increases the ability to count low ionizing radiations flux (low energy and activity (< 1 count /s). The setup can thus be used for long term studies, without detector derivation.

The diameter of the probe ranges from 1 to 5 mm and the length can be up to several tens of meters. The probes can be installed in non-human accessibility places like radioactive areas, underground galleries, wells and even can be inserted in industrial endoscopes... The use of optical fibers allows a maximum compactness of the probes, as well as human access to photodetectors for service and further evolution of the device.

#### Tracers for enhanced oil recovery in the Brazilian Northeast

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The use of tracer substances for petroleum reservoir characterization is growing a lot in the last years. The information obtained through this technology is crucial for the reservoir manager and they result in actions of great economical and environmental impact for the production of petroleum. The objective of this technology is to supply information concerning the distribution and path of the fluids involved in the reservoir of petroleum, defining the directional tendencies of flow and allowing the evaluation of the swept efficiency of the recovery processes. This application has as advantage to be the only tool that accesses the reservoir supplying direct information without causing physiochemical modifications to the system. Like this, a great effort has been made in the sense of developing methodologies for quantitative interpretation of tracer tests in petroleum reservoirs. In this sense, this work presents a tracer application for an advanced recovery petroleum pilot of the Brazilian Northeast. Two different tracers were used, one in the original condition of the area, where it would be applied the process, and the other, after the treatment. The obtained results demonstrated that the use of tracer is an indispensable tool for evaluation of recovery processes.

#### Using tracer data to improve petroleum reservoir models

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Numerical reservoir models are extensively used to plan and optimize production from petroleum reservoirs. In order to maximize the effect of enhanced oil recovery (EOR) methods and modern drilling technology (e.g. horizontal/flexible and multi-branched wells), it is important to obtain a proper understanding of the complex geological structures found in most reservoirs. Improved computational capabilities and simulation tools have helped to improve decisions regarding optimum reservoir development. Nevertheless, any reservoir simulation model represents one of many realizations of the real reservoir, and to reduce the inherent uncertainty of the models it is important to include additional data, such as tracer data. Unfortunately, tracers are underexploited as a source of data to understand oil reservoirs, and should be used in a better way, e.g., to locate and quantify bypassed and un-produced oil volumes in the reservoirs. In this paper we address this shortcoming, by illustrating how tracer data can be used in combination with other production data to improve reservoir models. Tracer modeling relies on high-quality tracer data that should always be evaluated for contamination, measurement flaws and misinterpretations. We discuss the issue of water re-injection, and demonstrate how this potential problem can be overcome. The value of tracer-data for simulation model improvements is demonstrated by history matching of a North Sea reservoir case using tracer and production data.

## Axial mixing performance of continuous phase in a pulsed sieve plate extraction column using gallium chloride as radiotracer

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Axial mixing performance of continuous phase in a liquid-liquid extraction pulsed sieve plate column has been investigated. Residence Time Distribution (RTD) analysis of continuous phase has been carried out for a wide range of pulsation frequency and amplitude in a liquid-liquid extraction pulsed sieve plate column operating with water as continuous and kerosene as dispersed phase using radiotracer technology. The column was operated in the emulsion region and <sup>68</sup>Ga in the form of gallium chloride eluted from a <sup>68</sup>Ge/<sup>68</sup>Ga generator was used to trace the continuous phase (water). Axial Dispersion Model (ADM) with open-open boundary condition and two point measurement method was used to model the continuous phase hydrodynamics. A comparison of experimental and model RTD functions and their respective Mean Residence Time (MRT) revealed that the Axial Dispersion Model is a suitable model to describe the continuous phase hydrodynamics in a pulsed sieve plate extraction column.

Keywords: Axial mixing; liquid-liquid extraction; pulsed sieve plate column; Residence Time Distribution (RTD); radiotracer; <sup>68</sup>Ga

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#### Groundwater recharge assessment using environmental tracing methods

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Groundwater resources are very important to the economic development in many areas, and sometimes is the only available one. Under natural conditions groundwater recharge is balanced by discharge, but this equilibrium is often affected by pumping. If the extraction of groundwater exceeds the amount of recharge, water level declines and consequently groundwater reservoir decrease. Besides, salt water intrusion from other reservoirs can take place and some groundwater dependent ecosystems can be influenced negatively. So, groundwater recharge and discharge need to be assessed in order to manage the aguifer system properly.

Environmental tracers can provide information related to the sources of groundwater and location of recharge areas and can be used to estimate recharge rates and the time of residence of groundwater. The oxygen and hydrogen isotopes (180, 2H) of the water molecules themselves are tracers of water movements and they are very useful to determine the sources, mechanism and seasonality of recharge and the interaction between surface water and groundwater. Chloride is a major ion that behaves conservatively and it is also a good tracer of groundwater movement. The mass balance of chloride is used to infer recharge mechanism and to estimate the magnitude of chloride addition to the aquifer. To identify the moment from which groundwater was recharged, some chronological tracers like 3H or 36Cl are used.

The objective of this work is to present the assessment of groundwater recharge using environmental tracers, particularly oxygen-18, deuterium, tritium and chloride. The study area is located in the center of the Santa Fe province, Argentina. It is a flat area with a slope of about 0.3% and the climate is moderate and humid. The conceptual model of the aquifer systems, previously defined, shows that it is a multilayer one. It is formed by an unconfined aquifer which is locally recharged from precipitation and a semiconfined one which received regional and local recharge.

The oxygen-18 and deuterium contents of groundwater in relation to the local meteoric water line allowed confirm that recharge comes principally from precipitation, it occurs during summer and falls, and no significant evaporation takes place during recharge process. The residence time of groundwater results in: no more than 5 years for the unconfined aquifer and more than 50 years for the semiconfined one, taking into account the tritium content.

An analysis of the chloride and stable isotopes contents of rainfall was done. The application of chloride mass balance method shows that recharge to the unconfined aquifer was approximately 63mm/year and the income regional flow to the semiconfined aquifer was 41mm/year. It is important to remark that although several important populations of the region are near to rivers, human activities are strongly dependant on groundwater. The semiconfined aquifer has been exploited for long time and nowadays the demand of groundwater is increasing quickly. The results of this work are very important and contribute to the knowledge of the groundwater system and provide quantitative criteria to base management and protection of groundwater resources and dependent ecosystem.

## Natural radioactivity of coastal sediments as "Tracer" in dynamic sedimentology

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Radiometric measurement of gamma natural radiation is a simple and fast technique for lithological mapping of the sea bottom that could provide useful information about the origin and transport of sediments.

- Natural radioactivity of sea bottom sediments can provide:
- granulometric selection of sediments,
- depth limit of wave effect on the sediments on the sea bottom,
- direction and distance of distribution of fluvial sediments,
- accretion and erosion zones along the coast line.

Natural radioisotopes distributed in sediments are uranium-238 (and its family), thorium-232 (and its family) and potassium-40.

The concentrations of U-238, Th-232 and K-40 in sediments vary with sediment nature and origin.

Based on these concentrations and decays, it can be calculated that the major contributor of gamma natural radioactivity of sediments is Th-232 (over 55%), thus Th-232 is the "natural tracer" of sediment dynamics. Th-232 is always in equilibrium and is very resistant against chemical and mechanical agents, so it makes it a reliable tracer for long period transport of sediments.

The scope of the study was to validate the natural radioactivity method for tracing sediment transport. Natural radioactivity investigation was undertaken in two important areas of the Adriatic Sea littoral of Albanian cost, in the gulfs of Durres and Vlora, where complex studies (radiotracers included) were carried out in the frame of the maintenance of existing Durres harbour, and the design of the new harbour in Vlora.

## <sup>115</sup>Cd/<sup>115</sup>In tracer generation development for industrial applications

Jussara Brant de Carvalho, Amenônia Maria Ferreira Pinto, Rubens Martins Moreira Gilmara Lúcia Souza Alvarenga, Fernanda Cristina Fonseca Camargo

Atualmente o paradigma é o gerador de 99Mo/99mTc sendo a medicina sua principal área de aplicação. Porém, estes geradores precisam ainda ser avaliados e validados para aplicações à indústria e ao meio ambiente. Este trabalho apresenta uma nova proposta: a produção de um gerador de radioisótopo de índio, baseado no par <sup>115m</sup>Cd/<sup>115m</sup>In. Isótopos de índio, como <sup>111</sup>In, <sup>113m</sup>In e <sup>115m</sup>In, têm sido amplamente estudados. Destes, <sup>113m</sup>In e <sup>115m</sup>Cd podem ser obtidos através de geradores. Para isto é utilizado como precursor o <sup>115m</sup>Cd, obtido através de irradiação de cádmio enriquecido em <sup>114</sup>Cd sob fluxo de nêutrons em reator nuclear.

Geradores de índio também podem ser constituídos pelo par 113Sn/113mIn. Embora a meia vida do <sup>115</sup>Cd (53,4 h) seja menor do que a do <sup>113</sup>Sn (115,1 d), o cádmio foi escolhido como radioisótopo pai devido à possibilidade de produzi-lo em reatores nucleares de pesquisa de pequeno porte a partir do <sup>114</sup>Cd isotopicamente enriquecido. O que se perde na vida útil de cada carga pode ser compensado com vantagem por se prescindir da importação do gerador. O <sup>115</sup>Cd foi obtido por irradiação de 0,01 g de <sup>114</sup>Cd, sob a forma de <sup>114</sup>CdO isotopicamente enriquecido a 98%, sob fluxo de nêutrons 6,6x1012. n/cm<sup>2</sup>.s, durante 5 horas.

Dois diferentes complexos de cádmio foram sintetizados em duas séries de experimentos: CdI<sub>4</sub><sup>2</sup>- e CdCl<sub>4</sub><sup>2</sup>-, sendo CdI42- o mais estável quimicamente entre os dois. As sínteses foram feitas pelo método Mirza<sup>[1]</sup> e Ehrhardt<sup>[2]</sup>. Antes de se utilizar o <sup>114</sup>CdO submetido ao reator, foram feitos testes preliminares da síntese destes dois complexos a partir de CdO não enriquecido ou radioativo. A formação dos complexos foi confirmada por análises de espectrometria de infravermelho.

Soluções de cloreto ou iodeto de cádmio, contendo a espécie <sup>115</sup>Cd, foram submetidas à cromatografia. Utilizou-se colunas de vidro, 1,3 cm de diâmetro por 20 cm de altura, empacotadas com 10 g de fase estacionária tamisada a 50 mesh. Foram testadas quatro fases estacionárias distintas: sulfato de zircônio, sílica gel, alumina G e dowex 1x8. ácido clorídrico foi utilizado como a fase móvel. O pH do eluente foi otimizado.

As frações recolhidas foram analisadas quanto a possíveis contaminantes arrastados mecanicamente pelo ácido clorídrico: zircônio, silício e alumínio quando presentes na fase estacionária. A adsorsão do 115Cd na coluna, bem como a formação de 115In foram acompanhadas por análises de espectrometria gama nas soluções antes de serem eluídas na coluna cromatográfica e nas frações recolhidas ao final desta.

Foram determinados os rendimentos da complexação, o grau de pureza do eluído com relação a AI, Si, Zr, bem como a retenção do <sup>115</sup>Cd/<sup>115m</sup>Cd pela coluna dowex 1x8 e o rendimento do filho <sup>115m</sup>In via espectrometria gama. A concentração e vazão do eluente foram otimizadas.

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## <sup>159</sup>Gd- Gadodiamide: obtainment of a possible therapeutic radiopharmaceutical

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The Gadodiamideâ(General Electric Healthcare Company), a non-ionic complex (Figure 1) of low dissociation in aqueous solutions (Chang et al., 1992). Actually it is the most used as a MRI contrast agent due to its low osmolality and chemotoxicity (Meyer et al., 1990).

The gadolinium present in Gadodiamide is a mixture of the isotopes 152Gd (0.20%), 154Gd (2.18%), 155Gd (14.80%), 156Gd (20.47%), 157Gd (15.65%), 158Gd (24.84%) and 160Gd (21.86%) (Browne, et al., 1986).

Submitting a natural gadolinium sample to an adequate neutron irradiation process, the radioisotope 159Gd is observed, which is a beta (1.001 keV) and gamma (main energy 363.54 keV) emitter (Moralles et al., 1995). According to some published studies, these energies are suitable for the diagnosis and therapeutic applications (Thrall et al., 2001).

The aim of this paper was to verify if the specific activity of 159Gd obtained by neutron irradiation would be suitable for its future use in radiopharmaceutical treatments.

#### Metodologia para aplicação de traçadores radioativos em reservatórios de petróleo

Alberto Avellar Barreto, Amenônia Maria Ferreira Pinto, Rubens Martins Moreira, Bruno Garcia Batista

O petróleo é considerado uma fonte de energia não renovável. É a matéria-prima da indústria petrolífera e petroquímica resultando em diversos materiais. Sabe-se que a taxa de recuperação do óleo contido em reservatórios de petróleo, caso não ocorra algum tipo de intervenção, é de aproximadamente 25%. Assim, utiliza-se a injeção de diferentes fluidos no reservatório com o objetivo de expulsar o óleo residual para fora dos poros em que estão retidos. Estas técnicas, podem elevar o coeficiente de extração para até 50% do volume original de petróleo no reservatório. O Centro de Desenvolvimento da Tecnológica Nuclear (CDTN) e a Petróleo Brasileiro SA (PETROBRÁS) vem realizando estudos em reservatórios de Petróleo por meio da aplicação de traçadores radioativos. As informações geradas visam aumentar a compreensão do comportamento do fluido de injeção, geralmente a água. Neste trabalho é apresentada a metodologia utilizada no CDTN para a aplicação de tracadores radioativos em reservatórios de petróleo. Esta metodologia foi desenvolvida seguindo os princípios de radioproteção definidos pela Comissão Nacional de Energia (CNEN). A injeção dos traçadores em pocos de petróleo é realizada por meio da utilização de cilindros amostradores. São descritos os procedimentos para a definição da quantidade de traçador a ser utilizado; para a preparação do esquema para a injecão do tracador; para o transporte do tracador até o local da injecão; para a execução da operação de injeção do traçador; para a execução da amostragem nos poços produtores; para a análise das amostras; e a elaboração de relatórios técnicos. Também são relatadas as vantagens em relação aos métodos utilizados anteriormente e as principais dificuldades enfrentadas ao longo da execução de todo o processo de injeção de traçadores em trabalhos já realizados pela equipe do CDTN.

## <sup>222</sup>Rn determination in water samples by using liquid scintillation spectrometry with different cocktails

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222Rn is a natural alpha emitter, which is produced in the reservoir rocks system and becomes partitioned in a welldefined proportion between the water and oil phases of the reservoir. The purpose of the present work is to develop an analytical methodology for the determination of 222Rn in surface water and in groundwater that will make possible the use of this radionuclide as a natural tracer in oil reservoirs. Liquid scintillation spectrometry is the technique most often used for determining 222Rn in water. Some measurement parameters had been determined for evaluation: scintillation cocktail, pulse shape analyzer (PSA), counting efficiency, counting region, spectral quench parameter of external standard (SQP(E)) and sample channels ratio (SCR). The 241Am, 90Sr and 226Ra standard solutions were used for the calibration of the spectrometer. The counting vial type was chosen in terms of energy resolution and cost. The quench effect and interferences due to other radionuclides in the measurement have also been studied. The sample was collected looking for minimizing radon leakage.

Keywords: Liquid scintillation Couting; Pulse Shape Analyzer; Tracer; Radon

## A non uniform flow velocity equation and its applications to mass transport in natural streams

A. Constain <sup>[1]</sup>, A. Carvajal <sup>[2]</sup> and J. Carvajal <sup>[3]</sup>

It is possible to find a more general equation for mean velocity of flow stating a thermodynamic relationship between hydraulic losses and macroscopic motion that drives the process. This new equation links the dispersion coefficient and advective mean velocity by means of a random walk definition using theories proposed by I. Prigogine in last years of XX century. This equation contains a state function that allows to know when the tracer has reached the "complete mixing" condition being a very powerful theoretical approach to study contaminant dynamic models. The researchers explain how they have developed a software tool that may be applied in real time stream studies.

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## Analytical model for tracer transport in formations having conductive geological faults

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Hydrocarbon recovery in oil reservoirs is importantly affected by the presence of geologic faults since they alter fluid motion. It becomes therefore very relevant for injection and flooding recovery projects to know with opportunity the presence of such heterogeneities and take their impact into account. Tracer tests provide means to dynamically characterize the conduit or barrier flow behavior of faults. Although diverse analytical models have been developed to describe tracer transport in geological conductive faults, only few of them capture the fact that injection and production wells are regularly located off the fracture plane, and take in consideration the tracer path covered outside the fault. This work proposes an analytical model to describe this situation. The system assumes three coupled regions in a bidimensional horizontal plane associated to the injector-to-fault, fault, and fault-to-production-well zones. The model introduces different uniform advection and hydrodynamic dispersion in each region. This last important phenomenon has not been considered in previous works. The initial condition is a Dirac delta pulse at the injection site. The equations are analytically solved in Laplace space and the inverse transform is evaluated numerically by the Stehfest algorithm. The sensitivity of the tracer breakthrough curve and the space pulse shape to fault length and fault dispersivity. Differences in the pulse profile and breakthrough curve are found in comparison to the case when the fault is not present. The developed model can be applied on tracer test interpretation in reservoirs having conductive faults.

Work submitted to the 5th International Conference on Tracer and Tracing Methods, to be held in Tiradentes, Brazil from 2 to 6 November 2008.

Application of radioactive tracers in pulp follow up during a digester blowing in a multiple batch-digester pulp mill

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The bottom blow pipelines at Arauco Pulp Mill Line 1 Plant presented severe damage at the blow valves in a repetitive fashion after performing a major process and piping modification in March 2007. The geometry of the blow tank, where top and bottom blow pipelines discharge gas and liquor from the digesters, was modified in such a way that top and bottom blow line flows faced each other in the circular tank. Several tests were performed on-site to determine the cause of the damages. In all tests, liquor appeared downstream of the valves in situations where the line should have been empty. Such phenomena could have been explained by choked flow in the lines or by crossing liquor flows at the blow tank head.

To determine the real cause of the appearance of liquor in the lines, an iodine-based radioactive tracer was added during the filling phase of one of seven digesters to its feed material so as to follow the flow of the outgoing liquor during the discharge phase. Sensors were placed at all bottom blow lines and at top and bottom blow inlets near the blow tank head so as to monitor possible crossing flows. This test was performed twice, injecting the tracer on two different digesters so as to measure the phenomena under slightly different circumstances. Gathered data showed the presence of crossing flows among the top and bottom blow lines, causing liquor to appear in pipelines that should have been free of any fluid. Choked flow was ruled out due to these results.

By performing two tests it was possible to determine that the phenomena was not restricted to the discharge of one digester vessel, but was associated to all seven digesters in the plant because of the geometric configuration of the nozzles at the blow tank. Due to the characteristics of the tracer used in the tests, it was possible to determine that top blow flows, which should be mainly gas, carried more liquid than expected. Also, by analyzing signal intensity at the discharge sensor it was possible to quantitatively determine the degree of homogenization of the mixing in the digester. In the end, crossing liquor flows were ruled as the cause for the problem in plant operation thanks to the application of tracing techniques, leading to a final design solution for blow tank and pipeline geometry.

#### Características hidrodinámicas del DSP de la Empresa Mielera "Heriberto Duquesne" por el método de los radiotrazadoresHydrodynamic Characteristics of SDU Enterprise "Heriberto Duquesne" by the radiotracer method.

Griffith J, Derivet M; Cuesta J, Flores J, Valdés J.

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Preliminary results achieved in the evaluation of the hydrodynamic characteristics of the settling dissolute unit (SDU) at the wastewater treatment plant located in the molasses enterprise "Heriberto Duquesne" working in continuous regime employing distillery slops diluted with water are presented.

Tc-99m was employed as tracer, its injection was performed instantaneously with the treated distillery slop fluid and the monitoring of the tracer through different areas of the unit was carried out with the help of radiation detectors. The control of specific parameters of the fluid as pH, conductivity, temperature, Brix (dissolved solids), Chemical Oxygen Demand (COD) and Settling Index was performed through the discrete sampling method.

Resuts achieved during the evaluation have shown that the pattern flow is far away from the theoretical one that is expected for a settling unit, the hydrodynamic model reflects the combination of a Tank in series flow with interchangeable "dead "zones due baffle design and the irregular fluid inlet to the system.

Recommendations are presented in order to eliminate the above mentioned deficiencies with the aim to increase the efficiency of the settling unit.

## Comparison of the PC, TLC-RP and HPLC-RP methods in the determination of the radiochemical purity of <sup>99m</sup>Tc-sestamibi

Almeida, E. V.; Fukumori, N. T. O.; Guirau, A.; Mengatti, J.; Matsuda, M. M. N.

Technetium-99m (99mTc) is the most frequently used radionuclide for in vivo imaging studies in Nuclear Medicine. Technetium-99m hexakis 2-methoxy isobutyl isonitrile (99mTc-sestamibi) is a monovalent, cationic, lipophilic complex that consists of one atom of 99mTc in a +1 oxidation state and six molecules of 2-methoxy isobutyl isonitrile (MIBI). It is a promising radiopharmaceutical for myocardial perfusion imaging and can be utilized in patients with acute myocardial infarction to quantify the amount of myocardium at risk. It is recommended at least 90% radiochemical purity (RCP) of 99mTc-sestamibi for clinical use. The present study reports the comparison among three chromatographic methods for determination of 99mTc-sestamibi purity: paper chromatography (PC), thin layer chromatography - reversed phase (TLC-RP) and high performance liquid chromatographic reversed phase (HPLC-RP), 30 minutes and 4 hours after reconstitution of the sestamibi lyophilized reagent with sodium pertechnetate (Na99mTcO4). Sestamibi lyophilized kit and generator eluate were from IPEN-CNEN/SP (Brazil). All reagents were from Merck (Germany). Water was purified in a MilliQâ system from Millipore (France). A labelling kit was reconstituted by the addition of 2 mL generator eluate containing 3.0 mCi Na99mTcO4 and heating for 10 minutes in a boiling waterbath. The PC mobile phase was obtained with equal volumes of acetonitrile, acetic acid 0.5 mol L-1 and sodium chloride 0.35 mol L-1 solution, while 10 volumes of tetrahydrofuran, 20 volumes of ammonium acetate 0.05 mol L-1 solution, 30 volumes of methanol and 40 volumes of acetonitrile were mixed for TLC-RP. A 5mL sample was applied both for the PC and TLC-RP methods. The analysis by HPLC-RP was performed with a Shim-Pack VP-ODS colunm (150 mm x 4.6 mm i.d., 5 mm particle size). A 25 mL aliquot sample was injected and the mobile phase (20:35:45; v/v, acetonitrile, ammonium sulphate and methanol, respectively) was isocratically eluted, at 1.5 mL min-1 flow rate, continuously monitored by a radiation detector Bioscan. Each kit was analyzed in duplicate in the three methods. The time after labeling of the kit was not significant for the RCP values (up to de 0.27 %). The values TcO-4impurity were  $(0.35 \pm 0.02)$ % by HPLC-RP and  $(1.86 \pm 0.20)$ % by TLC-RP. The TcO2 values were  $(0.52 \pm 0.02)$ % by PC and  $(0.34 \pm 0.02)$ % by 0.02)% by TLC-RP. PC and TLC-RP RCP were (99.48  $\pm$  0.09)% and (97.75  $\pm$  0.08)%, respectively. A HPLC-RP analysis showed a labeling average efficiency of  $(99.36 \pm 0.06)$ %. Each method gave similar results for radiochemical purity. The difference between HPLC-RP and TLC-RP was 1.73%, while the difference between HPLC-RP and PC was only 0.12%. The identity of some radiochemical impurities of the 99mTc-sestamibi was confirmed by 3 different methods. PC, TLC-RP and HPLC-RP techniques are important aids in quality control of <sup>99m</sup>Tc-sestamibi radiopharmaceutical.

#### Profile analysis of fluids displacement in a Phase separation tank applying the radiotracer technique

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This paper aims at studying and evaluating a phase separator tank (organic/liquid) applying the radiotracers technique. This technique is based on the study of material transport and identification of operational problems in industrial tanks and has great advantages due to the interventions done without influencing on the normal operation. In the studied case, to evaluate the fluid hydrodynamic behaviour inside the tank, the radiotracer 82Br in the aqueous solution of potassium bromide (KBr), and for the radiotracer passage registration, four scintillation detectors of NaI (2 x 2)" were used. For methodology was possible to analyze the unit response and also to identify the operational problems.

#### Determining tritium in new tracers research

Raquel M. Mingote, Eliane S. C. Temba and Rubens M. Moreira

Tritium is a radioisotope of hydrogen which disintegrates by emission of beta particle to 3He. Its determination is important in several applications, such as groundwater survey, quantification of aquifer recharge, and oil recovery from reservoirs, in which tritiated water is used to tag injection fluids. Since an oil field has many injection and production wells, it is necessary to use more than one tracer to discriminate the individual contribution of different injection in an production wells. The development of a variety of tracers for use in oil fields is an objective of the research group at CDTN.

Tritiated water, considered as an ideal tracer, is used as a reference for the analysis of the behavior of the new tracers in tests realized in reduced physical model of the reservoir tests aimed at verifying the occurrence of diverse interactions that may occur with the tracer during the investigation process. To determine tracer response the tritium content in the samples is measured by liquid scintillation spectrometry.

The present work presents the methodology employed in this analysis, improvements that have been made, and also those that are still required. A low background liquid scintillation system detector, Perkin Elmer-Wallac Quantulus 1220, has been used.

Due to the low volume of samples usually obtained in such tests, small polyethylene vials have been used. Since the sample characteristics varied with the characteristics of the tracer studied, the use quenching correction methods and changes in the sample load capacity were needed.

#### Determining tritium in new tracers research Radon determination in different matrices using LSC

Eliane S. C. Temba, Raquel M. Mingote and Amenônia M. F. Pinto

<sup>222</sup>Rn has been used as a natural tracer in several studies in hydrogeology, in assessment of residual NAPL contamination of aquifers and as tracer in oil fields. In this work it is presented methods developed for the determination of 222Rn in different matrices using Liquid scintillation counting (LSC). It were performed measurements in sandstone samples, to determine the radon emanation rate, and in liquid vaseline samples, to evaluate the viability of measuring radon in oily samples.

It was defined the optimum parameters of counting, like sample/scintillation cocktail ratio, the appropriated scintillation cocktail for each sample, the type of vial, the counting windows, etc.

In the case of sandstone samples it was developed a methodology to count solid samples by LSC, using a certified soil sample from IAEA to determine counting efficiency. The detection limit obtained was 2 Bq/kg with a counting efficiency of 66%. To the liquid vaseline samples it was used a reference standard solution of <sup>226</sup>Ra from NIST. Using an immiscible cocktail and a low diffusion polyethylene vial, it were attained a detection limit of 0,08 Bq/L and a counting efficiency of 88,9%, considering the 214Po counting region.

#### Development of new and alternative tracers for oil reservoirs

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Natural production mechanisms or primary production contribute to the extraction of about 25% of the original petroleum in the reservoir. This means that the greater part of petroleum is retained in the rock pores. Tracers are used in the exploitation of petroleum reservoirs in order to obtain information that help to optimize the yield of produced oil. One of the most conspicuous applications of tracers to reservoir exploitation is in the evaluation of secondary recovery techniques, due to their capability of providing information over an extended area, differently from other techniques that are restricted to discrete points.

Multiple tracer injection is often required and an option of interest is afforded by activable tracers. These can be fed to the system in the inactive state, be sampled collected production wells, and irradiated and measured by Neutron Activation Analysis. Some of the elements of the lanthanide series that exhibit very high thermal neutron cross sections have been tested for this purpose.

A properly performing tracer should not lag behind the liquid flow nor be lost through interactions with the rocks and non aqueous fluids inside the reservoir. Hence, they must be quite hydrophilic and have nearly null partition coefficients, so that they do not adsorb on the internal surfaces of the rock pores nor migrate to the oil phase. It is necessary to complex the lanthanide metals with appropriate ligands. A complexation methodology has been optimized. Its products were characterized and the yield quantified by 1H and 13C NMR, by Infrared Spectrometry and by Thermogravimetric Analysis (TGA)

Following that, the performance of these lanthanide complexes as tracers has been tested in essays that simulate the possibility their adsorption (Kd) in the solids inside the reservoir, by means of core tests. Tests have been carried witheuropium and dysprosium, that had been complexed with diethylenetriaminopentaacetic acid,(DTPA), and with thiacarboxylic acids the general formula R-S-Y-COOH. The core tests also displayed a good match between the lanthanide complexeds and the reference tracer used (tritiated water).

## Gamma-ray survey in the characterization of an area contaminated by mercury in Descoberto-MG, Brazil

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Following an intensive rain period occurred in December 2002, it was observed in the rural zone of the Municipality of Descoberto, State of Minas Gerais, Brazil, the arising of metallic mercury in the slope of the right margin of the Rico brook, an affluent of the Grama creek, inserted in the basin of Paraíba do Sul river. The site was characterized as Contaminated Area and the results of chemical analyses determined the interruption of the supply of public water and the interdiction of an area of about 8,000m2. According to local people reports and based upon historical reconstitution, it was found out that gold mining works were carried out in the region from the 19th century until early 20th century. The mercury used in the concentration of gold imposed a great environmental problem. In 2006 the Detailed Investigation of the site was concluded, showing that the highest mercury concentrations occurred in the gold gravels found along old structures known as "canoes", which were used to concentrate and depurate the gold. In this process, the miners used to throw the metallic mercury straight into the "canoes" in order to promote the amalgamation between gold and mercury. This product was then collected and burned, and the gold, recovered. Mercury is also found in soil, close to the gravel layers, but in lower concentrations than gravels. The Contaminated Area comprises about 800m2 and is characterized by mercury concentrations in soil and gravel higher than the proposed remediation limit, which is of 10mg/kg and was established by risk analysis. The contamination reachs no more than 1m deep into the soil sub-surface. The existence of the radioactive mineral monazite (Ce, La, Y, Th) PO4 as a constituent of the gravel allowed the use of a gamma detector (SRAT SPP2 Gamma Scintilometer) as an important tool for the determination of the spatial distribution of the gravel contaminated by mercury. This paper intends to present the results obtained by the radiometric survey realized in the Contaminated Area, so to demonstrate how this survey can be useful in investigating the aurific monazitic gravels occurring in the old gold mining works in the region of Descoberto/MG, which are potential sources of contamination by mercury. Reference (background) counting values for soils in the region ranged between 40 and 60cps (counts per second), while the higher value measured in the gravel was of 500cps.

#### Implementation and automatization of a flow-through experimental setup for the evaluation of new tracers for oil reservoir characterization

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Petroleum, nowadays, is one of the most important natural resources of the world, since it is one of the main primary sources of energy and also raw material of many different products. It can be found accumulated in the subsoil, and was produced from the remaining portions of animals and vegetables, in a process that took millions of years to be concluded. Initially, only a small fraction of the oil contained in a reservoir (about 25%) can be extracted, and for this reason water is injected in the deposit to increase the recovery of the remaining quantity. Aiming to optimize this process, called Secondary Recovery, tracers are used to provide information concerning the water flow inside the reservoir. In order to be applied as a tracer, a certain compound must fulfill a series of requirements, depending on the desired purpose. Tracers applied to study the water behavior inside the reservoir, must follow water molecules without sticking to the rocky surfaces. This phenomenon, named sorption, can be investigated in different ways, and the two most used methods are laboratory batch method and laboratory flow-through method. The latter has the advantage to incorporate hydrodynamic effects in its results. The present paper describes an automated experimental setup to study sorption characteristics of compounds synthesised in CDTN on consolidated sandstone samples from the Botucatu formation, in which motorized valves and a software for remote control and acquisition of data have been developed. The system performance was investigated using NaCl as reference tracer - since its behavior is in quite good agreement with that of tritiated water, which is considered the ideal tracer for water phases - and its concentration in the effluent was determined by direct conductimetry, by means of a flow-through cell at horizontal position, since it changes the hydrodynamic behavior of the compound when vertically positioned. Moreover, the pump outflow should be kept bellow 5 mL/min, otherwise the balance will not reach stability, thus corrupting the transmission of data to the computer. Finally, breakthrough curves from automatizated and manually controled experiments were compared, and their results were quite similar, confirming that the construction and automatization of the system for flow-through tests were successful.

## Preparation of radioactive Gold-nanoparticles as a SPECT imaging agent

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Nowadays, molecular imaging using radiopharmaceuticals has been a technique of choice for tumor diagnosis due to its higher resolution [Watanabe Y, Brain nerve 59 (3): 209, 2007]. Colloidal gold nanoparticles (AuNPs) represent a novel technology in the field of particle-based tumor-targeting drug delivery, early-stage diagnosis and tumor radiotherapy. Gold nanoparticles or colloidal gold have been assayed in mice as X-Ray contrast agent for tumor diagnosis [Cai Q-Y et al, Invest. Radiol. 42:797, 2007]. However, the amount of gold in this technique is high (2.7 g kg<sup>-1</sup> animal), becoming virtually impracticable for humans. In this study, we described our efforts to develop a radioactive AuNP probe with specific activity high enough to serve as an agent for single photon emission tomography (SPECT) imaging using just a few amount of gold (~ 10 mg kg<sup>-1</sup> animal). In order to prepare the AuNPs, chloroauric acid solution (HAuCl<sub>4</sub>) was reduced by sodium citrate and nanoparticles diameter was determined by atomic force microscopy (AFM) and optical absorption. AuNPs around 3 nm were produced and submitted to irradiation at TRIGA Mark IPR-R1 reactor for 8 hours, under a thermal and epithermal neutron flux of 6.6 x  $10^{11}$  and 3.0 x  $10^{10}$  n cm<sup>-2</sup> s<sup>-1</sup>, respectively. Under this flux the reaction <sup>197</sup>Au(n, g) <sup>198</sup>Au occurred producing radioactive gold nanoparticles. The induced activity obtained for the <sup>198</sup>AuNPs was around 800 mCi/mg<sub>Au</sub> at the end of the irradiation. Radiochemical analysis was done by gamma spectrometry using a germanium detector. A surfactant agent, polivinylpirrolidone (PVP K30), was mixed to the AuNPs to keep the homogeneity of colloidal gold in physiological solution and to avoid further agglomeration. AFM showed diameters of 60-80 nm for PVP coated colloidal gold (AuNPs-PVP).<sup>198</sup>AuNPs-PVP was intravenously injected in Swiss mice and toxicology was evaluated at different periods post injection. The <sup>198</sup>AuNPs-PVP proved to be biocompatible without significant toxicity in the mice. Toxicological results state for the safe use of these nanoparticles for imaging diagnosis. Studies are ongoing to evaluate the potential use of <sup>198</sup>AuNPs-PVP as blood-pool SPECT imaging agent.

Keywords: <sup>198</sup>Au Nanoparticles, SPECT agent, toxicology

Financial support: FAPEMIG, CNEN, CNPq, CAPES

#### Radiolabeling and quality control of 2-acetylpyridine N4-phenyl- thiosemicarbazone: a potent antitumoral agent

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The neoplasms basic knowledge is increasing quickly, however, few advances have been reached in clinical therapy and diagnosis of tumours. Application of radiotracer (radiopharmaceutical) highly specific for tumours constitutes a non invasive approach for early diagnosis of malignant tumours by molecular imaging. Therefore, the development of alternative radiopharmaceuticals is relevant in the attempt to improve prognosis and to increase the patient survival. N4-phenyl-2-acetylpyridine thiosemicarbazone (Ph) presents a wide range of bioactivities including antitumoral activity; nonetheless, its property as a radiopharmaceutical is still unknown. For this purpose, Ph must be radiolabeled with high specific activity and radiochemical purity. The aim of this work is to radiolabel Ph using 125I as radiotracer and to carry out the quality control of radiolabeled molecule. Labeling was done by lactoperoxidase method and 1251-Ph specific activity and radiochemical analyses were done using ascending chromatography in Whatman paper nº 1 as the stationary phase and methanol saturated with KI as mobile phase. Contaminants, mainly as 125I-, were eliminated by anionic exchange chromatography. 125I-Ph serum stability and interaction with blood plasmatic proteins were also analyzed. 125I-Ph production was successful with 96% of radiochemical purity and high specific activity (17.6 TBq /mmol). 125I-Ph showed to be a stable compound keeping its stability for 7 days, when stored at 2-4°C. Moreover, 125I-Ph was found to be stable at 37°C in serum for at least 4 hours. The low binding to plasmatic proteins (7.9±1.1%), during 24 hours of incubation, suggests that 125I-Ph can freely circulate in the blood stream, without main interferences on its biodisponibility. 125I-Ph proved to possess indispensable characteristics for a possible antitumor radiopharmaceutical to be applied for non invasive tumour diagnosis. The next step will be to evaluate 1251-Ph biodistribution and its capacity for tumour detection in vivo.

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## RTD of the gas phase in a 130 m<sup>3</sup> self-aerated flotation cell

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In flotation processes the gas flowrate (typically air) is a key variable which provides the gas surface required for selective mineral particles capture and transport. For this purpose the gas flowrate must be firstly dispersed into small bubbles and the bubble swarm contacted with the pulp (mineral suspension) in order to collect the mineral particles of interest. As a result particle-bubble aggregates are generated, which are less dense than the medium, thus allowing the separation of collected minerals by true flotation. Measurement of the gas residence time distribution (RTD) is relevant because it allows the evaluation of the gas mean residence time as well as the effective gas content in the cell. Presence of gas recirculation through the rotor and gas short-circuiting to tails can also be identified.

In this work the direct measurement of the gas residence time distribution in an industrial 130 m<sup>3</sup> self-aerated flotation cell is presented. For this purpose a radioactive tracer gas was produced in the 5 MW Nuclear Reactor from Chilean Nuclear Energy Commission.

The gas selected was Freon 13B1, which contains Br for activation with a 36 hours mean life, which was compatible with the required times for preparation, activation, manipulation, transport and application of the gas tracer in the industrial flotation process.

The gas tracer was injected as an impulse signal at the gas (air) inlet point, located at the top of the cell, from which the gas tracer circulates first through the rotor, where the bubble dispersion occurs, and then the gas becomes well distributed over the whole cross-section area before leaving the cell.

The transient response curves obtained from these experiences showed that the air entering the cell was preferentially circulated around the upper half of the cell. Also, it was found that the presence of circulating gas near the bottom of the large size flotation cell was not significant, therefore that gas short-circuiting was negligible.

## Scintillating setup for low activity beta emitter detection in underground galleries

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We have developed a new family of miniaturized setups for the detection of ionizing radiation (alpha, beta, gamma, X). The device consists in a plurality of high accessibility scintillation probes. Each probe is made of a scintillating crystal optically coupled to several optical fibers. The outputs of these fibers are detected in coincidence by photomultipliers: a light pulse emitted by the crystal will be validated if photons are detected by each photomultiplier simultaneously. This selection allows considerable reduction of the effect of the "dark counts" of the photodetectors and thereby increases the ability to count low ionizing radiations flux (low energy and activity (< 1 count /s). The setup can thus be used for long term studies, without detector derivation.

In this particular application, the particles to detect a beta emitter tracer (36Cl, Emax 710 KeV). The aim of this experimentation is to study diffusion in geological clay. The system including photodetectors and electronics will be installed in the underground gallery, and the probes will be inserted in 20 m long, 20 mm wide drillings in the clay. This setup allows accessibility to all the detection system during the 5 year long study which is needed for recalibration, service and control.

This poster will describe the technical configurations of this experiment and the associated advantages.

#### Soil-plant transfer factors of typical Brazilian crops

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Phosphogysum (PG) or agricultural gypsum (AG), a solid waste from the phosphate fertilizer industry, is used as a soil amendment, especially in soils from the Cerrado region, in Brazil. Nevertheless, such material may contain natural radionuclides and metals which can be transferred to soils, plants and water sources. This paper presents and discusses the results of physical and chemical analyses that were done in order to characterize samples of PG and two typical soils from the Cerrado region, Brazil, one clayey and one sandy. These analyses included: solid waste classification, evaluation of organic matter content, P, K, Ca, Mg, and Al concentrations, particle size distribution, and mineralogical composition. Natural radionuclides and metal concentrations in PG and soils samples were also measured. According to Brazilian Regulation-ABNT Norm 10004/2004, the results showed that PG is classified as non-inert solid waste. The soils studied presented high acidity and low natural fertility. It was also observed that the activity concentration of natural radionuclides in PG was below the limit suggested for agricultural purposes. In addition, this study verified that natural radionuclides and metal concentrations in PG were lower than those concentrations verified in the clayey oxisol from Sete Lagoas, Minas Gerais State, Brazil. Soil-plant transfer factors of natural radionuclides were determined for three different types of crops: lettuce, soybeans and corn. For this, a set of greenhouses experiments were carried out during a period of one year. The obtained values varied from 4.7 E-04 for <sup>238</sup>U (corn) to 9,6 E-02 for <sup>210</sup>Pb (soybeans).

## Synthesis of the indium complexes for using as an activable tracer in the oil secondary recuperation

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Oil is found in the environment, impregnated in the sedimentary rock pores: the reservoirs. Such reservoirs are layered of natural gas, hydrocarbonates and water, submitted to high pressure. The perfuration of a well can push the oil off the rock pores to the surface this process is called primary recuperation In this case, it is obtained approximately 25% of the existing oil. The rest is obtained by a process named secondary recuperation, in which a fluid is injected in order to dislocate the oil towards the well of production. The tracer is used to evaluate the secondary recuperation of the oil and in this present paper studied tracer is Indium

Keywords: Activable Tracer, Indium

#### Tracer techniques as a contribution for studying the hydrological behavior of a São Francisco River sub-basin - PART I

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A research program that has been developed by CDTN/CNEN, since 1997, in order to study the hydrological behavior of a São Francisco river sub-basin is presented in this paper. This program involves a series of research projects which main objectives are to use tracer techniques, available in CDTN/CNEN, to increase the knowledge on the hydrological behavior of a typical basin of the Minas Gerais State central region as well as to improve, or to adapt, methodologies that, although developed in basins with different characteristics from the Brazilian basins, have been used in our country to quantify hydrological phenomena. A summary of three already developed research projects and their results are presented here. The paper was divided in three parts, one for each research project. In this first part, it is presented the project entitled "Application of Tracer Technique to Flood Studies in the Juatuba Representative Basin", which had as main objectives the determination, with the tracer technique, of the Time of Concentration and the Instantaneous Unit Hydrograph - basic parameter in flood studies - and to evaluate the empirical formulas used in Brazil to estimate these parameters.

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## Tracer techniques as a contribution for studying the hydrological behavior of a São Francisco River sub-basin - PART II

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#### Tracer techniques as a contribution for studying the hydrological behavior of a São Francisco River sub-basin - PART III

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A research program that has been developed by CDTN/CNEN, since 1997, in order to study the hydrological behavior of a São Francisco river sub-basin is presented in this paper. This program involves a series of research projects which main objectives are to use tracer techniques, available in CDTN/CNEN, to increase the knowledge on the hydrological behavior of a typical basin of Minas Gerais State central region as well as to improve, or to adapt, methodologies that, although developed in basins with different characteristics from the Brazilian basins, have been used in our country to quantify hydrological phenomena. A summary of three already developed research projects and their results are presented here. The paper was divided in three parts, one for each research project. In this third and last part, it is presented the project entitled "Study of the Runoff Formation using Natural Tracers in the Juatuba Representative Basin", which has the following objectives: i) to test the methodology that uses the tracer technique to separate the surface and groundwater runoff parcels of flood hydrographs; ii) to test the use of natural tracers, as electric conductance and isotopes (180 and 2H), to make the runoff separation; and iii) to establish a comparison between the results obtained with this methodology and the results that would be obtained by the graphical methods.

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