New Perspectives for the TRIGA IPR-R1 Research Reactor

A. S. Leal, R. C. O. Sebastião, R. R. Rodrigues, M. A. B. C. Menezes

Nuclear Technology Development Centre/CDTN – Brazilian Nuclear Energy Commission/CNEN Av. Antônio Carlos 6627 - 31270-901, Belo Horizonte, MG, Brazil *E-mail address of main author: asleal@cdtn.br*

K. Krambrock

Department of Physics, Universidade Federal de Minas Gerais/ UFMG Av. Antônio Carlos 6627 - 31270-901, Belo Horizonte, MG, Brazil

E. Pereira-Maia, W. Guerra, F. C. S. de Paula

Department of Chemistry, Universidade Federal de Minas Gerais/ UFMG Av. Antônio Carlos 6627 - 31270-901, Belo Horizonte, MG, Brazil

Abstract. This work presents the recent results of the new applications using the TRIGA IPR-R1 reactor of the CDTN. Labeling of special compounds to be used in biological and pharmacokinetic investigations ; coloring of Brazilian gemstones irradiated by neutrons and preliminary results obtained from a neutron beam using the MCNP code are presented.

INTRODUCTION

The TRIGA IPR-R1 CDTN's research reactor is of the type MARK I, which core is below of the floor level, as it is shown in Figure 1. It is operating since 1960 and its main activities have been the neutron activation analysis and the training of nuclear power plant's operators. Since 2001, new projects of utilization of the reactor were initiated as the production of some special labeled molecules to be used in medical purposes and the improvement of color of Brazilian gemstones by neutron irradiation [1,2,3].



FIG. 1. View of the well of the TRIGA IPR MARK 1 reactor [4].

More recently, new projects to evaluate the possibility of obtaining a characterized vertical neutron beam from a vertical tube from the TRIGA's core was initiated. The availability of a neutron beam with appropriate characteristics as intensity, spectrum and collimating, will enlarge the possibilities of applications in these fields [4]. This work presents the recent results of these new applications, about gemstones, special activated molecules and also the preliminary simulated results of the neutron extractor using the MCNP code.

PRODUCTION OF RADIOLABELLED COMPOUNDS

During the last five years, researches involving new drugs or new strategies of drugs administration opened a very interesting and important field of application for the TRIGA reactor: the labeling of special molecules to be used in biodistribution and for pharmacokinetic studies *in vitro* and *in vivo*. The first compound irradiated in the TRIGA with this purpose was the CDDP, cis-dichlorodiammineplatinum (II), Pt(NH₃)₂Cl₂. CDDP is an effective chemotherapeutic largely used to treat systemic tumors in several organs: testicles, ovary, head, neck, bladder. However, its side effects are serious principally for the nephrotoxicity [5-9]. Investigation of new formulations containing CDDP to minimize or eliminate these effects is extremely relevant. A detailed description of irradiation of CDDP by CDTN and its applications are described elsewhere [2].

Besides CDDP, in the last years new compounds of platinum and other metals with antitumoral and/or antibiotic activity has being investigated as the tetracycline-platinum II (Tc), $Pt(C_{22}H_{24}N_2O_8)Cl_2$ complex . Tetracycline is one of most important antimicrobial agents with broad spectrum, but its low toxicity, low cost and oral administration have inducing an indiscriminate use with a consequent appearance of bacterial resistance. Previous studies performed with Tc, showed a more pronounced antibacterial effect compared to tetracycline [10] and more recent results [2] showed also an enhancement of the antitumoral activity of the radiolabeled Tc. Further *in vivo* studies must be performed to confirm this important potential use of Tc. Once a chemotherapy agent is radiolabeled it is interesting to investigate the possible synergic effect of its chemotherapy and radiotherapy actions. The preliminary results obtained using the radiolabeled platinum compounds of in *vitro* studies showed very interesting results [2]. A full description of this work will be sent to publish briefly.



FIG. 2 Molecular structures of CDDP (left) and tetracycline platinum [15].

STUDY OF BRAZILIAN GEMSTONES BY NEUTRON IRRADIATION

Brazil is one of most important miners of gemstones in the world but most of the them is colorless or present only weak blue coloration and therefore, has very low commercial value. In this work, preliminary tests including different gem minerals, colorless topaz, diamond and spodumene were irradiated by neutrons with a flux of 4×10^{12} n/cm²s and an integrated dose of 10^{17} and 10^{18} neutrons/cm² corresponding to 7 and 70 hours of irradiation time. Initially, the main objective was the observation of coloration, measurement of the total activity and the procedure to determine activated

Alexandre Soares Leal

trace elements. After irradiation the samples were measured by gamma spectroscopy. Samples that presented activation until three times of the background limit were considered for the analysis by optical absorption and electron paramagnetic resonance (EPR).

Figure 3 shows three examples of gems treated by neutron irradiation: topaz; spodumene and diamond. All samples were colorless or very pale colored before irradiation. Spodumene and diamond did not show any activated elements; topaz dependent on the origin of the samples: from Marambaia, MG, showing only signals due to ⁴⁰K; from Tocantins, TO, in addition ⁴⁶Sc and from Rondônia, RO, ⁴⁶Sc and ⁵¹Cr. Potassium isotope ⁴⁰K has lifetime of about 1.43 x 10⁹ years and originates in general from natural irradiation. The ⁴⁶Sc isotope results from the reaction ⁴⁵Sc(n, γ)⁴⁶Sc and has a lifetime of about 28.1 days and ¹³⁴Cs from the reaction ¹³³Cs (n, γ)¹³⁴Cs with lifetime of 2.06 years. The excited state of Cs is metastable, ^{134m}Cs, with lifetime of 2.9 hours. The ¹²⁴Sb isotope results from the reaction ¹²³Sb (n, γ)¹²⁴Sb.

Differently from other gem minerals, which can be treated with gamma radiation, the induced color in topaz is either not stable or very weak. However, irradiation by neutrons result in medium to strong blue colors depending on dose. In this work, topaz from different regions in Brazil were irradiated with variable time of irradiation from 3.5h to 69.5h under a thermal flux of 4.0×10^{12} or 6.4×10^{12} neutrons/cm²s depending on the position of irradiation. Except for the samples of one specific region of the country, Minas Gerais, all remaining topaz samples presented a very low final activity allowing the work of characterization of defects and color centers induced by irradiation. Samples were analyzed by neutron activation using the neutron activation analysis method for impurities, by optical absorption for their color and by electronic paramagnetic resonance for radiation-induced color effects. The results obtained showed that the blue color induced by neutrons is independent of the origin of topaz and correlated with an O⁻ defect. Its concentration and the blue color are increased linearly by dose and both still not saturated for the highest applied dose [3].



FIG. 3 (Left) Topaz from Rondônia, RO, Brazil; (Centre) spodumene from the east of Minas Gerais, MG, Brazil and (Right) diamond from Minas Gerais, MG, Brazil after neutron irradiation with dose of 10^{18} , 10^{17} and 10^{18} neutrons/cm².

At present, neutron irradiation in Brazil for commercial use is prohibited by law, and all topaz treatments are done in foreign countries. The results obtained here can open new economic perspectives for the country in the international gem trading of topaz.

SIMULATION OF A VERTICAL NEUTRON BEAM

Simulations of the neutron flux along the vertical extractor in the TRIGA IPR-R1 research reactor were carried out by the MCNP-4B code [11]. The model adopted in the simulation reproduced the experimental setup used as a neutron extractor for a short time twenty years ago. The vertical tube consists of a 0.2 cm thickness, 525 cm long and 10 cm diameter aluminum cylinder [12] and experimental thermal and epithermal neutron flux measurements were performed using ¹¹⁵In foil. To The calculate the neutron flux, a model of the tube was inserted in the core reactor input file previously developed by Dalle [13]. Only its extremity surface in contact with the reactor core is closed and the vertical extractor is filled with air. The Figure 2 shows the view of the neutron beam and the core of the reactor. Table 1 shows the experimental and calculated results for the thermal flux in the positions 1, 2 and 3 of the tube as showed in the Figure 4.



FIG. 4. Lateral cut-away view of the TRIGA reactor model (left) and the top view (right above). Materials regions are specified in the figure. The point detectors along the vertical extractor are: 1 for 120 cm, 2 for 240 cm and 3 for 360 cm.

		Thermal neutron flux		(n/cm^2s)
Extractor	Position h (m)	Experimental	Calculated	Relative Error (%)
1	1,20	2,00 x 10 ⁸	1,0 x 10 ⁹	6
2	2,40	2,80 x 10 ⁷	1,0 x 10 ⁵	14
3	3,60	9,1 x 10 ⁶	5,0 x 10 ⁵	15

Table 1. Experimental thermal and thermal neutron flux the simulated results.

Alexandre Soares Leal

These preliminary results can be considered very positive with an acceptable range of relative error. Along the vertical tube the neutron fluxes were evaluated by point detectors with 1.5 cm diameter, placed in the positions 1, 2, 3 and 4, as illustrated in Figure 3. Inside the neutron extractor, the neutron flux available permits the samples be irradiated and studied [14]. The thermal neutron flux close to $1,0 \times 10^7 \text{ n/cm}^2\text{s}$ in the superior extremity of the tube could allow new applications of utilization in the TRIGA.

CONCLUSION

The results presented here confirm that even a lower research reactor can be very useful and important alternative tool in different fields of research, as it is the case of new radiolabeled molecules and investigations of improvement of color in precious stones. The vertical beam IPR-R1 research reactor was calculated. The results obtained for the flux in the vertical beam showed a good agreement with experimental values obtained previously. The expected thermal neutron flux of 1.0×10^7 neutrons/cm² s in upper extremity of the vertical tube suggests that new applications, as doping of silicon, can be carried out in the next future. The availability of a neutron flux with a characterized energy spectra motivates the implementation of new fields of research using the reactor.

ACKNOWLEDGEMENTS

The authors would like to thank the CNPq and FAPEMIG financial support and express their special thanks to the IPR-R1 TRIGA operator team for their help during the experiments.

REFERENCES

- [1] IAEA-TECDOC-1234, 2001. The applications of research reactors, Report of an Advisory Group meeting held in Vienna, 4-7 October 1999.
- [2] Leal, A.S., Carvalho Jr., A.D., Abrantes, F.M., Menezes, M.A.B.C., et.al., Production of the radioactive antitumoral cisplatin, Applied Radiation and Isotopes, 64: 178-181 (2006)
- [3] Leal, A.S., Krambrock K., Ribeiro, L.G.M., Menezes, M.Â.B.C., Vermaercke, P., Sneyers, L., Nucl. Instr. and Meth. A, Study of neutron irradiation-induced colors in Brazilian topaz, to be published (2007).
- [4] Leal, A.S., Menezes, M.A.B.C., Vermaercke, P., Sneyers, L., Jensen, C.E.M., Investigation of chemical impurities in formulations, phytotherapics and polyvitaminic medicines by k0-instrumental neutron activation analysis, Nucl. Instr. and Meth. A ,564: 729-732 (2006).
- [5] Calvert, H., Judson, I., Van der Vijgh, W.J., Cancer Sur., 17:189 (1993).
- [6] Daugaard, G., Abildgaard, U., Cancer Chemother. Pharmacol., 25: 1 (1989).
- [7] Reynolds, J.E.F., Martindale the Extra Pharmacopeia, 513 (1999).
- [8] Stewart, C.F., Hampton, E.M., Am. J. Hosp. Pharm., 46: 1400 (1989).
- [9] Trissel, L.A., Martinez, J.F., Am. J. Health-Syst Pharm., 53: 1041 (1996).
- [10] Chartone-Souza, E., Loyla, T.L., Buciarelli-Rodriguez, M., Menezes, M.A.B.C., Rey, N.A., et-al, Journal of Inorganic Biochemistry, 99: 1001 (2005).
- [11] Briesmeiter, J. F., et. al., 1997. MCNP-4B A General Monte Carlo N-Particle Transport Code-Version 4B, Los Alamos National Laboratory, LA-12625-M.
- [12] Costa, A.L., Análise das aplicações de um extrator de nêutrons para o reator TRIGA IPR-R1, CNEN/CDTN, NI-IT4-008/02, Belo Horizonte, julho 2002.
- [13] Dalle, H. M.. Simulação do Reator Triga IPR-R1 Utilizando Métodos de Transporte por Monte Carlo, Ph.D thesis, Faculdade de Engenharia Química, UNICAMP, São Paulo (2005).
- [14] Neto A.J.D., Haddad C.M.K., Pelosi E.L., Zevallos-Chavez J.Y., Yoriyaz H., et. al., Monte Carlo simulation as an auxiliary tool for electron beam quality specification for intraoperative radiotherapy, Brazilian Journal of Physics 35-3B: 801-804 (2005).