2009 International Nuclear Atlantic Conference - INAC 2009
Rio de Janeiro,RJ, Brazil, September27 to October 2, 2009
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN
ISBN: 978-85-99141-03-8

RADIOGENIC LEAD-208 ABUNDANCE 88.34 %

José A . Seneda[1], Alcídio Abrão[1], Mauro S. Dias[1], Key Sato[2] , Mauricio H. Kakazu[1], Vera L. R. Salvador[1], Carlos A. S. Queiroz[1], Soraya M. R. da Rocha[1], Walter R. Pedreira[3]

[1]Instituto de Pesquisas Energéticas e Nucleares - IPEN-CNEN/SP
Av. Lineu Prestes, 2.242
05508-900 São Paulo, SP
jaseneda@ipen.br

[2] Universidade de São Paulo Instituto de Geociências
Rua do Lago, 562
05508-900 São Paulo, SP

[3] Fundação Jorge Duprat Figueiredo- Segurança Méd. Trabalho
Rua Capote Valente, 710
05409-002 São Paulo, SP

ABSTRACT

Brazil has a long tradition in thorium technology, from the monazite ores mining until the production of the nuclear grade thorium compounds. Early in 1969 the Institute of Energy and Nuclear Research (IPEN) designed a project for a pilot plant installation to purify the thorium compounds, based on the solvent extraction technique. Thorium compounds used came from monazite´s industrialization. During the course of the operation of this plant, a crude sludge were formed containing thorium not extracted and the whole rare earths, plus minor impurities like sodium, titanium, zirconium, hafnium, iron, silicon, phosphate and the thorium daughters were accumulated. Included is the radiogenic lead-208. This sludge, hereafter named "RETOTER", was treated with hydrochloric acid and the lead was separated and recovered by anion exchange technology. The lead-208 was analyzed by mass spectrometry (HR-ICPMS) technique. The lead-208 abundance measure was 88.34%, this allowed the calculation of the thermal neutron capture cross section of $\sigma^0\gamma$ = 14,6 +/- 0.7 mb, considerably lower than the $\sigma^0\gamma$ = 174.2 +/- 0.7 mb value of the natural lead.

1. INTRODUCTION

Brazil has been produced Thorium nitrate, nuclear grade, from monazite ore, in a pilot plant at Instituto de Pesquisas Energéticas e Nucleares (IPEN-CNEN/SP). The extraction process by TBP/Varsol solvent produced a raffinate, about 28 ton, containing impurities of Iron, lead and rare earths. From this raffinate it was separate the lead by anionic exchange column, this has a valuable increase in the isotope Pb-208 (88.34%) from the decay of Th-232.

Actually the lead isotope Pb-208 is produced by physic process, magnetic enrichment or centrifugation, turning expensive product [1]. The Pb-208 demand is world wide increasing due to new technologies, specially in the nuclear reactor Accelerator Driven System [2] which use this isotope as cooling and target besides is using in the fusion experiments of new elements [3].

Seneda et al. presented the process to separated and obtained lead from the rafinate in a nitrate thorium pilot plant using anionic exchange technique [4].

The scope of this work is to show the measurement of the Pb-208 by mass spectrometry technique using the High Resolution Ion Coupled Plasma Mass Spectrometry HR-ICPMS.

2. MATERIALS AND METHODS

The separation and obtaining process of isotopic lead, consist in the dissolution of rare earth hydroxide and impurities containing lead, in hydrochloric acid which come from a pilot plant solvent extraction for the thorium purification follow by a exchange anionic column [4-8].

The lead eluted from anionic resin was used to isotopic analysis, with concentration adjusted to 1.0 $mg.L^{-1}$ in Pb, that were determined by HR-ICPMS and Thermal Ionic Mass Spectrometry TIMS.

The HR-ICPMS instrument used in this work is a Finnigan MAT, model ELEMENT, (Bremen, Germany). This instrument presents a double-focusing ion analyzer, with reverse geometry (Nier-Johnson): a magnetic analyzer is followed by an electrostatic analyzer and can operate in three nominal resolution values: 300, 3000 and 7500. Thermoinonic Mass Spectrometer (TIMS), Micromass VG isotopes Mod. 354 with HP system – software VG ISSUE III.

Analytical grade acids (Suprapure from Merk, Germany) and purified water produced using a Milli Q (from Millipore, France) system, were used for dilutions.

In order to reduce the risk of contamination (of external origin or cross contamination between the samples), extremely severe measures were adopted to assure that materials used during the sampling execution were sufficiently cleaned and that all the sample handling and processing were conducted in extraordinary clean ambient.

In order to determine the thermal neutron capture cross section for heavy Pb, the values of cross section for isotopes ^{204}Pb, ^{207}Pb and ^{208}Pb were taken from the literature [9]. From the isotopic abundance for the natural Pb and the abundance in the present work for heavy Pb, the average thermal neutron capture cross sections were calculated.

3. RESULTS AND DISCUSSION

The results of isotopic analysis presented in Tables 1 and 2 and Fig. 1 shown the average obtained in the both Thermionic Mass Spectrometry (TIMS) and HR-ICPMS for Pb natural and heavy Pb from elution of chromatographic resin. These results shown, the increase of heavy Pb in ^{208}Pb, due to raw material that contain ^{232}Th, that decay to ^{208}Pb.

Table 1.– Abundance and Thermal Neutron Capture Cross Section (σ_γ^o) of Natural Pb

Natural Pb		
Isotope	Abundance %	σ_γ^o [11] (mb)
204	1.40	661. +/- 70.
206	24.10	30.6 +/- .8
207	22.10	712.+/- 10.
208	52.40	0.49 +/- 0.03
	σ_γ^o Pb nat =	**174.2 +/- 7.0 mb**

These results permitted to calculated the thermal neutron capture cross section (see Table 1 and Table 2), an important parameter for nuclear technology.

The thermal neutron capture cross section (σ_γ^o) from this lead has been obtained using the values of the neutron capture cross section of the isotopes Pb-204, Pb-206, Pb-207 e Pb-208 in the literature [9].

Table 2 – Thermal Neutron Capture Cross Section (σ_γ^o) of heavy Pb

Heavy Pb		
Isotope	**Abundance %**	σ_γ^o **[11] (mb)**
204	**0.44**	**661. +/- 70.**
206	9.97	**30.6 +/- .8**
207	1.18	**712.+/- 10.**
208	88.34	**0.49 +/- 0.03**
	σ_γ^o Pb =	14.6 +/- 0.7 mb

For "heavy" Pb this value corresponds to only 8.5% when compared to the natural Pb, that can be explained because ^{208}Pb has the smallest capture cross section among the Pb isotopes and appears in "heavy Pb" with highest isotope abundance (see Table 1 and Table 2).

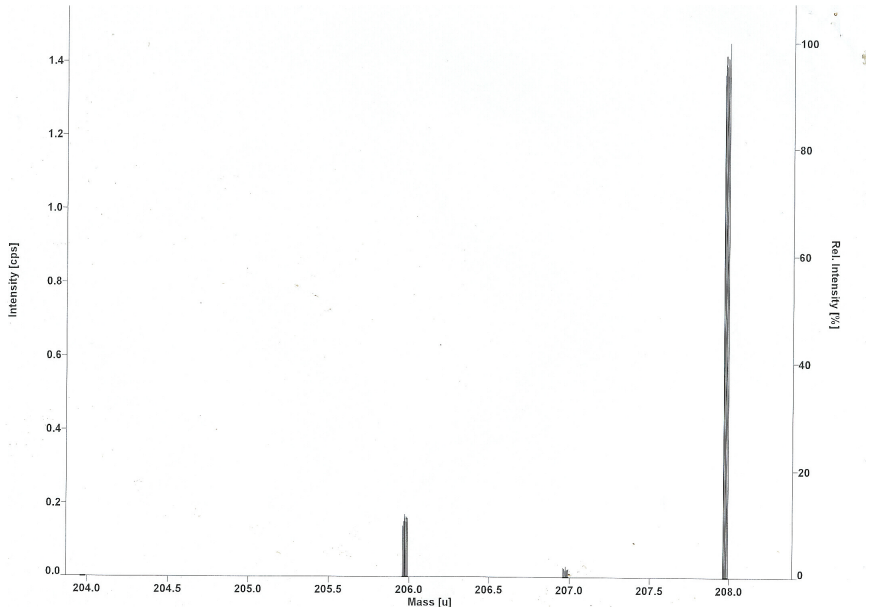

Figure 1: HR-ICPMS Spectrum from "heavy" lead.

4. CONCLUSIONS

In this work the lead-208 was analyzed by mass spectrometry (HR-ICPMS) technique. The lead-208 abundance measure was 88.34%, this allowed the calculation of the thermal neutron capture cross section of $\sigma^0\gamma$ = 14.6 +/- 0.7 mb, considerably lower than the $\sigma^0\gamma$ = 174.2 +/- 0.7 mb value of the natural lead. The "heavy" Pb obtained in this process is an important raw material to be considered in nuclear technology because of the different isotope abundance in ^{208}Pb and lower cross section that justify its recovery. This production of the isotope ^{208}Pb, nowadays has a considered interested by increased uses in the synthesis of new elements and as tracer in several applications.

ACKNOWLEDGMENTS

The authors wish to thank, Eng. Alvaro Ikuta, Dr. Oscar Vega and Dr. Jorge E. S. Sarkis.

REFERENCES

1. Production of Stable Isotopes http://www.tracesciences.com/links.html (2009).
2. Neutronic And Physical Characteristics Of An Accelerator Driven System With A Target And Core Coolant From The Lead-208 Isotope http://accelconf.web.cern.ch/accelconf/r06/HTML/TUEO.HTM (2009).

3. P.J. Karol, H. Nakahara, B. W. Petley, E. Vogt, On the discovery of the elements 110-112. *Pure Applied Chemistry*, **v. 73**, ed. 6, pp. 959-967 (2001).
4. J.A. Seneda, C.A.L.G. de O. Forbicini, C.A. da S. Queiroz, M.E. de Vasconcellos, S. Forbicini, S.M. da R. Rizzo, Vera L.R. Salvador, A. Abrão, Study on radiogenic lead recovery from residues in thorium facilities using ion exchange and electrochemical process, *Progress in Nuclear Energy*, ***In Press, Corrected Proof***, *Available online 12 May* (2009).
5. K . J.Bril, P. Krumholz, Developments in thorium production technology**,** *Proceedings of the Third Internacional Conference on the Peaceful Uses of Atomic Energy* New York, NY, Aug 31- Sept 9, 1964, New York :1965 : United Nations, **Vol. 12**, pp. 167-172 (1965).
6. K.J. Bril, P. Krumholz, Produção de óxido de tório nuclearmente puro. Report, Instituto de Pesquisas Energéticas e Nucleares-CNEN/SP, IEA-115 (1965).
7. A. Ikuta, Tecnologia de purificação de concentrados de tório e sua transformação em produtos de pureza nuclear. Estudo do sistema $Th(NO_3)_4 – HNO_3 – NaNO_3 –$ TBP – Varsol. Instituto de Pesquisas Energéticas e Nucleares-CNEN/SP (Master Thesis), (1976).
8. D. Brandão Fº , E.C.B. Hespanhol, S. Baba, L.E.T. Miranda, J. A. Araújo, "Reaproveitamento do tório contido em resíduos provenientes da usina de purificação de tório**"** Report, Instituto de Pesquisas Energéticas e Nucleares –CNEN/SP,IEA-035 (1977).
9. S. F. Mughabghab, *Neutron Cross Sections. Neutron Resonance parameters and thermal cross sections*. Orlando, F.L. Academic Press. Inc , **Vol.1** part B, pp 82-1–82-29 (1984).
10. P.J. Karol, H. Nakahara, B. W. Petley, E. Vogt, On the discovery of the elements 110-112. *Pure Applied Chemistry*, **Vol. 73**, ed. 6, pp. 959-967 (2001).