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NUCLEAR FUEL MATERIALS ANALYSIS USING THERMAL NEUTRON CAPTURE GAMMA RAYS

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SAO PAULO

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NUCLEAR FUEL MATERIALS ANALYSIS USING THERMAL
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ABSTRACT

Concentrations of impurities or burnable poisons in reactor fuel materials which are not possible to detect with conventional methods of analysis were determined by measuring the prompt gamma rays following thermal neutron capture, in a reactor internal geometry. The calibration of the experimental arrangement was performed with UO_2 targets containing controlled amounts of Gd, Sm and Dy.

ANÁLISE DE MATERIAIS COMBUSTÍVEIS NUCLEARES USANDO RAIOS GAMA
DE CAPTURA DE NÊUTRONS TÉRMICOS

RESUMO

Pela medida dos raios gama prontos da captura de nêutrons térmicos de um reator, numa geometria de alvo interno, foram determinadas, em materiais combustíveis nucleares, concentrações de impurezas ou de venenos propositais que não podem ser detectadas por métodos convencionais de análise. A calibração do arranjo experimental foi feita com alvos de UO_2 contendo quantidades controladas de Gd, Sm e Dy.

INTRODUCTION

The interest of the man for the energy available from the atom has increased after the actual world energy crisis. Thus, in the design of nuclear power plants, special care has been devoted to the improvement of techniques of purification and analysis of the nuclear materials. Some typical contaminants of those materials are the rare earths like Gd, Sm, Dy and Eu. The burnable poisons are also of great interest. A burnable poison is a material with a high neutron absorption cross section (Gd, Sm, Dy, etc.) distributed in the core region of a reactor in such a way as to control its reactivity. Thus it is interesting to have a selective and nondestructive analytical technique capable of measuring the contaminants and the burnable poisons concentrations.

There exist already several techniques which are quite sensitive for the determination of trace impurities present in any material. However some of them, like spectrographic analysis, loose accuracy when measuring quantities greater than 30ppm. Neutron activation analysis relies on the properties of the product nucleus and in numerous cases the product nucleus may have a short half-life, a long

half-life or a stable composition or it may not be a gamma emitter at all. In the specific case of nuclear materials (like uranium, for example), the neutron activation analysis of the impurities turns difficult by the production of fission fragments in the matrix.

In a thermal neutron capture reaction, the gamma rays of the compound nucleus emitted within a very short time interval are the so-called prompt gamma rays and are detected simultaneuously with the sample irradiation. Prompt gamma-ray spectrometry offers advantages over many of other techniques in that it provides a trace and major multielement analysis requiring a minimum of physical preparation and no chemical processing. As prompt gamma-rays are typical for each element, the analysis of the spectrum obtained for a certain sample permits the identification of its components.

The main advantage of this technique is based on the relatively low neutron binding energy ($\sim$ 4.8MeV) of nuclear fuel materials which allows that all high energy capture gamma-rays belonging to the impurities can be detected in a relatively low background conditions. This occurs, for instance, in uranium matrices where typical rare earths contaminants or burnable poisons are easily detected due to their usual high cross sections and also high energy prompt gamma-rays.

Using a reactor internal geometry we have measured samples of uranium oxide (UO_2) containing controlled amounts of the rare earth impurities Gd, Sm and Dy. The prompt high energy gamma-rays of these elements were easily seen, since above 4.8MeV there are no more prompt gamma-rays emitted by the uranium matrix.

Sensitivity of the Method

The sensitivity of the method for the analysis of impurities using the prompt neutron capture gamma-rays may be seen in the following way:

If R is the capture rate, we have

$$R = \frac{mN_o}{A} \phi \sigma$$

Where: N_o — Avogadro's number

ϕ - Neutron flux (neutrons/cm^2 s) on the target

m,A,σ - mass (grams), atomic weight and neutron cross section (cm^2) for the specific isotope.

Since mass and flux are not physical properties of the isotope, we can consider that the capture rate is dependent only of the σ/A ratio. However, this ratio is the indication for the detection sensitivity of a given isotope only. Since we have a different emission probability for each gamma transition and also an isotopic abundance (f) of each isotope in the natural element, a better indication of the sensitivity (S) for each element is the product between the σ/A ratio, the emission probability and the isotopic abundance. The emission probability is given as gamma-rays per 100 captured neutrons (γ/100 captures) and is the so-called gamma-rays intensity[1].

The relative sensitivity S is then calculated as

$$S = I \frac{\sigma}{A} f$$

In Table I are presented some values of the relative detection sensitivity S calculated for several elements. Nuclear data like σ, A and f are from the Amersham-Searle "Chart of the Nuclides" (1968) and we used the Rasmussen[4] gamma-ray intensity and energy values. Calculations were made for the gamma ray of highest intensity of each isotope.

Table I

Relative Detection Sensitivity for Natural Elements with High Energy Gamma Rays

Elements	A	Z	$E\gamma$ (keV) (strongest transition)	$I\sigma/A$	$(I\sigma/A)f$
Be	9	4	6810	6.24×10^{-4}	6.24×10^{-4}
C	12	6	4945	1.9×10^{-4}	1.88×10^{-4}
N	14	7	5267	1.45×10^{-3}	1.45×10^{-3}
Na	23	11	6395	5.92×10^{-3}	5.92×10^{-3}
Al	27	13	7724	1.74×10^{-3}	1.74×10^{-3}
Ti	48	22	6760	8.91×10^{-2}	6.59×10^{-2}
V	51	23	6517	1.82×10^{-2}	1.81×10^{-2}
Ni	58	28	8999	3.45×10^{-2}	2.34×10^{-2}
Cd	113	48	5824	4.09	5.02×10^{-1}
Sm	149	62	7213	2.11	2.91×10^{-1}
Eu	151	63	5918	7.7×10^{-3}	3.68×10^{-3}
Gd	157	64	6750	21.0	3.3
Dy	164	66	5607	4.41×10^{-1}	1.24×10^{-1}
Ta	181	73	5965	8.63×10^{-4}	8.63×10^{-4}
W	183	74	6191	4.99×10^{-3}	1.32×10^{-3}
Hg	199	80	5967	1.58	2.67×10^{-1}
Pb	207	82	7368	3.25×10^{-3}	7.35×10^{-4}
U	238	92	4060	8.37×10^{-4}	8.31×10^{-4}

As can be seen from Table I, the probability of detecting impurities is strongly dependent on the neutron capture cross section. For example, the relative detection sensitivity for [58]Ni is low, despite the relatively high values of the gamma transition intensity (41.65%) and isotopic abundance (67.77%) because the neutron capture cross section is only 4.9 barns. On the other hand, [157]Gd has a high sensitivity because, even with a 1.32% gamma ray intensity and a 15.68% isotopic abundance, one has 250.000 barns for the neutron capture cross section. However, in multielement analysis of a complex material by nondestructive methods, it is difficult to define the detection limits uniquely, since they strongly depend on the experimental conditions and on the matrix nature which can present inter-element interference.

EXPERIMENTAL

a. Experimental Facilities and Targets

The experimental set-up for the analysis was installed in the lower tangential tube of the IEA-R1 reactor of São Paulo and the basic features of the internal target facility are illustrated in Figure 1.
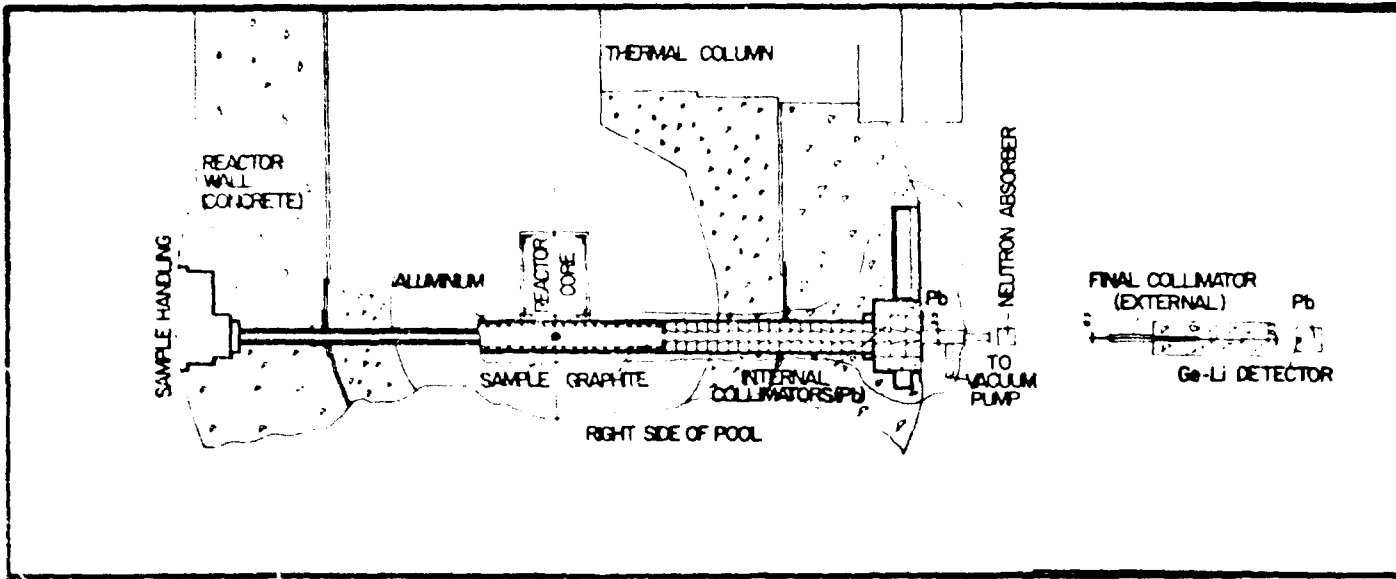
Figure 1 — Experimental Arrangement Installed at the tangential Tube of the IEA-R1 Reactor.

The internal target geometry is definitely superior to the external one because of the higher signal-to-background ratio. In the external target geometry there are two solid angles: one for the neutron at the target and other for the gamma rays at the detector, whereas in the internal geometry there is only one solid angle involved, from the target to the detector. So, despite the difficulty in handling the samples, the internal target geometry was chosen.

The samples to be analysed were positioned close to the reactor core in a thermal neutron flux of 3.7×10^{12} neutrons/cm^2.s and were seen by a single Ge(Li) espectrometer (an ORTEC true coaxial detector with an active volume of 42.5 cc) situated outside of the reactor biological shield. A ^{6}Li$_2$CO$_3$ thermal neutron absorber was installed in front of the Ge(Li) detector to protect it against probable thermal neutrons existing in the gamma ray beam.

In order to reduce the background, the following precautions were taken: a) The lead collimators inside the aluminium tube are conical behind the target in order to prevent the radiation from the beam tube wall close to the core reaching the detector directly; b) since gamma rays from the aluminium tube can be scattered at the target, the reactor beam tube, on the whole inner surface where there are no collimators, was covered with a tube of $3\frac{7''}{8}$ internal diameter, of nuclear pure graphite, a material emitting very few thermal neutron capture gamma-rays; c) the reactor beam tube was evacuated to less than 10^{-1} torr in order to decrease the scattered neutrons reaching the Ge(Li) spectrometer as well the capture gamma-rays from the nitrogen in the air; d) the target holder was made from nuclear pure graphite due to its low capture cross section and rather simple gamma spectrum. Also, the part of the target holder seen by the spectrometer had as little material as possible (1 mm thick).

A self-powered neutron flux detector of ^{104}Rh placed near the target in the reactor pool was used to monitor the total number of neutrons on the target. A typical fluence for the measurements was 3.2×10^{17} neutrons/cm^2.

The samples were prepared in the following manner: the impurities and the uranium oxide (UO$_2$) were carefuly weighted using a Mettler analytical balance. After the weighing, the components were mixed in an agate mortar until reaching homogeneity.

The calibration of the experimental arrangement was performed with uranium oxide (UO$_2$) targets with controlled amounts of gadolinium (Gd$_2$O$_3$), dysprosium (Dy$_2$O$_3$) and samarium (Sm$_2$O$_3$). The compositions of the used samples are presented in Table II. The total weight of the samples was around 5 grams.

Table II

Measured Samples

Sample	ppm of impurity	UO$_2$ mass (g)	Impurity mass (oxide) (mg)
UO$_2$ + Dy$_2$O$_3$	379.63	4.5834	1.997
	191.70	4.5436	1.0
	105.28	4.5498	0.55
UO$_2$ + Gd$_2$O$_3$	48.32	9.0030*	0.501
	45.09	4.5021	0.234
	22.85	5.0766	0.134
	13.28	5.0452	0.067
UO$_2$ + Sm$_2$O$_3$	296.56	4.4982	1.547
	129.15	4.5141	0.676
	70.18	4.5312	0.369
	37.4	4.4652	0.194

(*) From that quantity we prepared two samples containing the same 48.32 ppm of gadolinium but 4.4862 g and 4.5031 g of uranium oxide, respectively.

b. Data Analysis

A gamma-ray spectrum measured with a lithium-drifted germanium detector consists of a series of narrow peaks on a continuous background. The impurities identification is done through the peak energies and by means of the peak intensities the element amount is determined

For spectra from Ge(Li) detectors the best gamma-ray energies and intensities are often obtained by manual calculation. However, since these spectra often contain more than 50 peaks, manual selection of the peaks is not practical when large number of spectra are analyzed, so computer programs are used. The data analysis was performed by the GAUSS V program[2], in operation in our computer center. In this program, the peak is represented by a gaussian function and the underlying spectral background by a linear function

The basic mathematical operation is the nonlinerar least-square fit by which the parameters of the gaussian (height, width and position) are determined. For the energy calibration[5] we used the quite well known capture gamma-rays of nitrogen[1]. The peak position is used for the gamma-ray energy determination and the gamma-ray intensity is obtained through the peak area.

RESULTS AND DISCUSSION

Typical capture gamma-ray spectra of uranium oxide matrices containing the rare earth impurities are shown in Figures 2, 3 and 4. As one can easily see, above 4.5 MeV, only the impurities peaks are present

The impurity content is proportional to the peak area (intensity) and is expressed in ppm. The calibration curves obtained with the experimental arrangement of Figure 1 are shown in Figure 5. The impurity content in the uranium oxide expressed in ppm is plotted against the peak area ratio of the impurity to the uranium (strongest lines). For furthe details of the procedure see reference 3.

The reproducibility of the sample preparation method was verified by measuring samples containing gadolinium as impurity and prepared from the same batch. The observed deviation was less than 2%. Another test of reproducibility was obtained by relating the strongest uranium – 239 transition peak area with the uranium oxide target mass and the neutron fluence and the results showed a deviation less than 3%.

The reproducibility and the rather convenient sensibility for impurities content greater than 20 ppm permits to say that the prompt gamma-ray analysis method acts as a complementary method to the existing techniques.
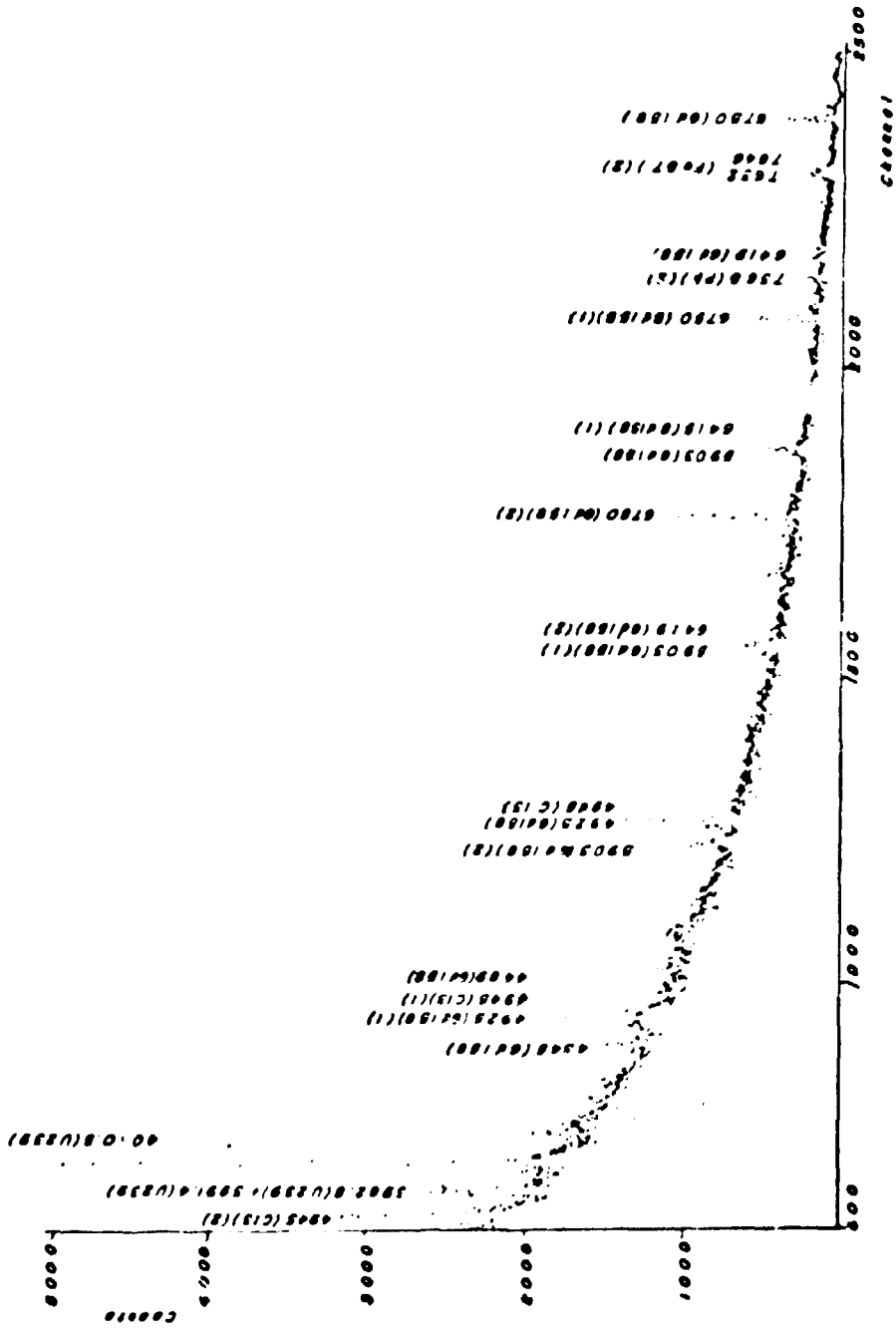
Figure 2 — Prompt Gamma-ray Spectrum of the (UO₂ + 379 ppm Dy) Sample in the Region of 3700 to 5800 keV. (1) and (2) are the Single and Double- — escape Peaks. Run of 25 h.

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Figure 3 — Prompt Gamma-ray Spectrum of the (UO₂ + 296 ppm Sm) Sample in the Region of 3900 to 6900 keV. (1) and (2) are the Single and Double-escape Peaks. The Iron and Lead Impurities are from the Graphite Target Holder. Run of 18 h.

Figure 4 — Prompt Gamma-ray Spectrum of the (UO₂ + 48 ppm Gd) Sample in the Region of 3900 to 6900 keV. (1) and (2) are the single and Double-escape Peaks. The Iron and Lead Impurities are from the Graphite Target Holder. Run of 20 h.
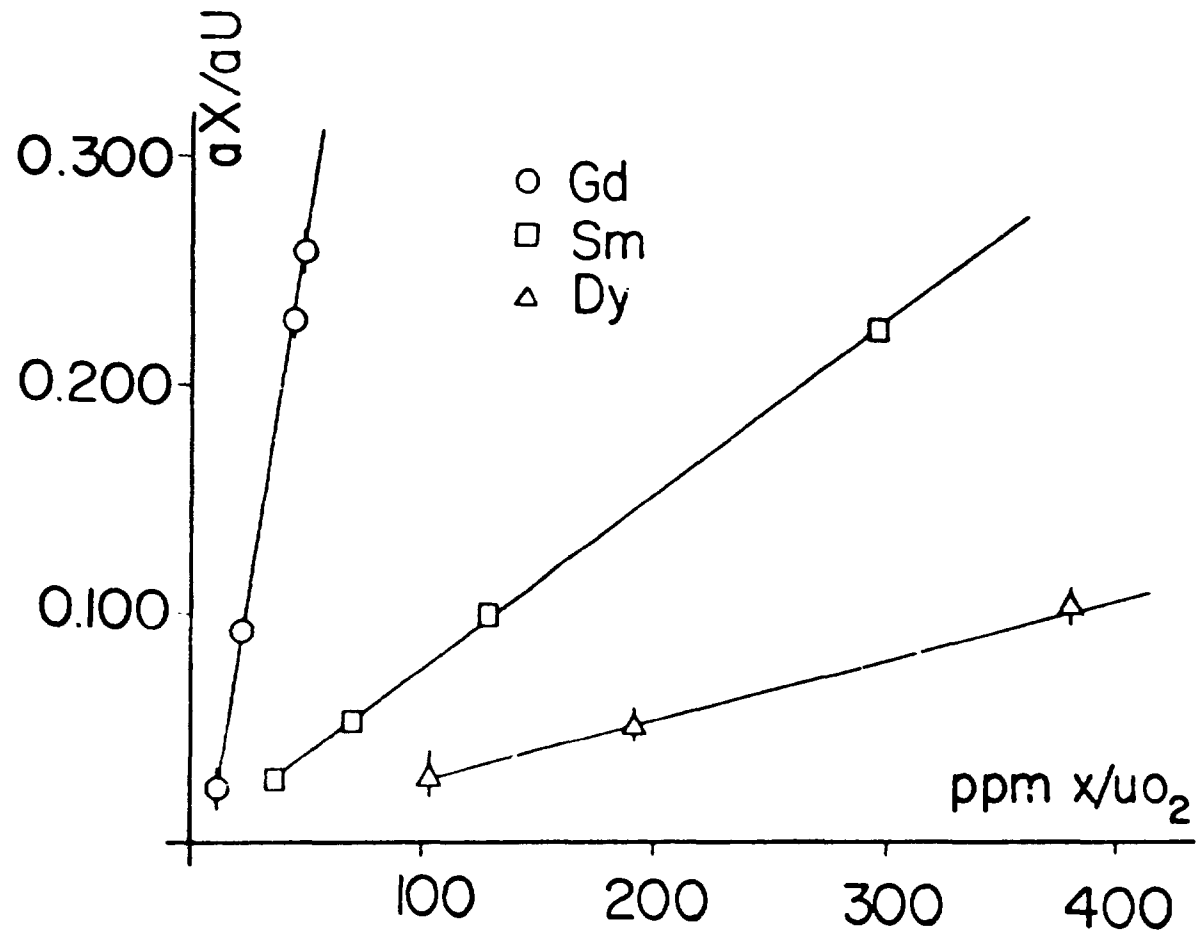
Figure 5 — Calibration Curves of the Experimental Arrangement. (ppmX/UO_2 : Impurity Content in Uranium Oxide; aX: Peak Area (double-escape) of the Impurity (5749.8 keV – [158]Gd, 7213 keV – [150]Sm 5606.7 keV – [165]Dy); aJ: Peak Area (full energy) of the 4060.5 keV of [239]U).

REFERENCES

1. AJZENBERG-SELOVE, F. Energy .vels of light nuclei A = 13 – 15. *Nucl. Phys. A*, 268:138, 1976.

2. HELMER, R. G. & PUTNAM, M. H. *GAUSS V, a computer program for the analysis of gamma-ray spectra from Ge(Li) spectrometers*. Idaho Falls, Aerojet Nuclear Company, Jan. 1972. (ANCR-1043).

3. PECEQUILO, B. R. S. *Nova técnica para a determinação de impurezas em compostos de urânio e de tório pela análise dos raios gama de captura*. São Paulo, 1977. (Tese de doutoramento). (IEA-DT-078).

4. RASMUSSEN, N. C. et alii. Thermal neutron capture gamma ray spectra of the elements. (AFCRL-69-0071). In: DUFFEY, D.; ELKADY, A.: SENFTLE, F. E. Analytical sensitivities and energies of thermal-neutron-capture gamma rays. *Nucl. instrum. Meth.*, 80:149-71, 1970

5. STOPA, C. R. S. & PECEQUILO, B. R. S. *Energy and relative efficiency calibration of a Ge(Li) gamma-ray spectrometer*. São Paulo, 1980. (IPEN-Pub-12).