

# Thermal neutron detection with a pyroelectrical detector: theory and results

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## Abstract

In this paper we present a theoretical model to explain the results obtained with a system used to measure thermal neutron fluxes. The detector system is constituted of a  $U_3O_8$  pellet with 20% enrichment in  $U^{235}$  attached to a pyroelectric ceramic. The thermal neutron fluxes was periodically modulated with a cadmium chopper. The energy released in the uranium fission heats the pyroelectric detector and produces an electrical signal directly proportional to the thermal neutron flux. The theoretical model shows also that it is possible to determinate the thermal diffusivity of nuclear fuels and the percentage of  $U^{235}$  enrichment in uranium samples.

## 1. Introduction

New methods to measure X-radiation energy fluence rate and the energy fluence of a brief exposure of diagnostic X-rays using pyroelectric dosimeters have been proposed since 1984 [1-4].

In 1992, S. B. Crestana et al. proposed a new technique for thermal neutron detection using pyroelectric ceramics [5]. The detector system is basically constituted of a lead zirconate titanate (PZT) ceramic attached to an uranium ( $U_3O_8$ ) pellet with 2 mm thickness. The energy released in the uranium disk gives rise to an electrical signal in the transducer. Thermal neutron fluxes within the interval from  $10^3$  to  $10^6$  n/(cm<sup>2</sup>s) have been detected using a  $U_3O_8$  pellet with 20% enrichment in  $U^{235}$ .

The detector response was shown to be directly proportional to the thermal neutron fluxes and inversely proportional to the chopper modulation frequency. In this paper we present a theoretical model to explain this results. The theory developed shows that the pyroelectrical signal is proportional to the sample enrichment in  $U^{235}$ .

## 2. Theoretical model

The background for our model is the Rosencwaig-Gersho theory [6] and the theory proposed by Mandelis and Zver for photothermal spectroscopy of solids [7]. Figure 1 shows the sample-transducer system. The backing is an aluminum plate. The PZT and the aluminum present a low cross section for thermal neutron reactions. We define the following parameters:

K: the thermal conductivity,

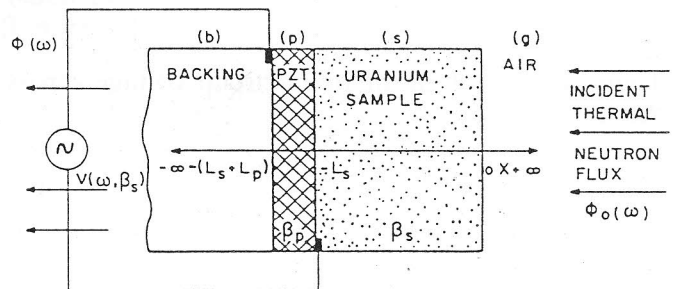


Fig. 1 - Schematic diagram of the neutron-pyroelectric detector system.

$\rho$ : the density,

$c$ : the specific heat,

$\alpha = K/\rho c$ : the thermal diffusivity,

$a = \left(\frac{\omega}{2\alpha}\right)^{\frac{1}{2}}$ : the thermal diffusion coefficient,

$\mu = \frac{1}{a}$ : the thermal diffusion length,

T: the temperature,

C: the capacitance of the pyroelectric element,

L: the thickness,

$\beta$ : the absorption coefficient,

p: the pyroelectric coefficient of the transducer.

We denote the following subscripts:

s for the sample

p for the pyroelectric ceramic

b for the backing

g for the air

The uranium sample ( $U_3O_8$  or  $UO_2$ ) is in the form of a disk having diameter D and thickness  $L_s$ . It is irradiated with a thermal neutron flux  $\Phi_0$ . The

sample absorption coefficient  $\beta_s$  varies with the sample enrichment in  $U^{235}$ . The interaction of thermal neutrons with the  $U^{235}$  produces nuclear fission, releasing the energy  $E$  as heat that propagates to the pyroelectric material. When the detector is heated its polarization changes. This change in polarization appears as a voltage  $V(\omega_o, \beta_s)$  on the capacitor formed by the pyroelectric material and the two electrodes. The temperature of the pyroelectric detector changes sinusoidally due to the modulation of the neutron beam. Due to the pyroelectric effect, the average surface charge on the electrodes of the detector can be expressed by:

$$\begin{aligned} \langle Q \rangle &= p A \langle \Delta T \rangle \\ &= (pA/L_p) \operatorname{Re} \left[ \int_{\text{esp } L_p} T(x) e^{i\omega_o t} dx \right] \end{aligned} \quad (1)$$

The average voltage in the pyroelectric detector is given by:

$$V = \langle Q \rangle / C \quad (2)$$

Then we can obtain the open-circuit voltage across the detector:

$$V(\omega_o) = \left[ \frac{pL}{K\epsilon_o} \vartheta_p(\omega_o) \right] e^{i\omega_o t} \quad (3)$$

where

$$\vartheta_p(\omega_o) = \frac{1}{L_p} \int_{\text{esp } L_p} T_p(\omega_o, x) dx \quad (4)$$

$T_p(\omega_o, x)$  is the temperature field inside the pyroelectric detector. We make the following assumptions:

- i)  $\beta_p = 0$
- ii) The neutron scattering is negligible
- iii) There is no heat loss in the sample detector boundary and in the lateral of the sample.

Then we can write the heat diffusion equations for the air, uranium sample and pyroelectric detector:

$$\frac{d^2}{dx^2} T_g(\omega_o, x) - \left[ \frac{i\omega_o}{\alpha_g} \right] T_g(\omega_o, x) = 0; \quad x \geq 0 \quad (5a)$$

$$\begin{aligned} \frac{d^2}{dx^2} T_s(\omega_o, x) - \left[ \frac{i\omega_o}{\alpha_s} \right] T_s(\omega_o, x) &= \\ = -(\Phi_o \beta_s / 2k_s) \exp(\beta_s x); \quad -L_s \leq x \leq 0 \end{aligned} \quad (5b)$$

$$\begin{aligned} \frac{d^2}{dx^2} T_p(\omega_o, x) - \left[ \frac{i\omega_o}{\alpha_p} \right] T_p(\omega_o, x) &= 0 \\ - (L_s + L_p) < x < -L_s \end{aligned} \quad (5c)$$

The pyroelectric detector is thermally thick. There is no heat diffusion to the backing. To specify the solution of (5a), (5b) and (5c), the appropriate boundary conditions can be obtained from the requirement of temperature and heat flux continuity at the boundaries. Then we obtain:

$$T_g(\omega_o, x) = c_1 \exp(-\sigma_g x) \quad (6a)$$

$$\begin{aligned} T_s(\omega_o, x) &= \left[ \frac{\Phi_o E \beta_s}{2K_s(\sigma_s^2 - \beta_s^2)} \right] \exp(\beta_s x) + \\ &+ c_2 \exp(\sigma_s x) + c_3 \exp(-\sigma_s x) \end{aligned} \quad (6b)$$

$$T_p(\omega_o, x) = c_4 \exp(\sigma_p x) \quad (6c)$$

where:

$$\sigma_j = -(1 + i) a_j \quad (7)$$

and  $V(\omega_o)$  can be expressed by:

$$V(\omega_o) = |V(\omega_o)| \exp \left\{ i \left[ \omega_o t - \Phi(\omega_o) \right] \right\} \quad (8)$$

Following the classification given by Rosenzweig and Gersho, we can classify the pyroelectric detector as optically transparent ( $\beta_p = 0$ ) and thermally thick ( $L_p \gg \mu_p$ ), and the uranium sample as semi-transparent for neutrons ( $\mu_{\beta_s} > L_s$ ) and thermally thick ( $\mu_s \ll L_s$ ).

### 2.1. Thermally thick and optically transparent detector

We define:

$$b_m = \frac{K_m a_n}{K_n a_n}; \quad r_j = \frac{\beta_j}{\sigma_j} \quad (9)$$

In this case there is no heat diffusion in the detector-backing boundary and the temperature equation is given by:

$$V(\omega_o, \beta) = \frac{p\Phi_o E}{2K\epsilon} \left[ \frac{\beta_s}{k_s(\beta_s^2 - \sigma_s^2)\sigma_p} \right] \left[ \frac{2(b_{sg}r_s + 1) - [(r_s + 1)(b_{sg} + 1)e^{\sigma_s L_s} + (r_s - 1)(b_{sg} - 1)e^{-\sigma_s L_s}]}{(b_{sg} + 1)(b_{ps} + 1)e^{\sigma_s L_s} + (b_{sg} - 1)(b_{ps} - 1)e^{-\sigma_s L_s}} \right] e^{-\beta_s L_s} \quad (10)$$

The equation (10) shows that the pyroelectric signal depends on the thermal parameters and on the interactions of neutrons with the uranium sample.

2.2. Uranium sample semi-transparent for neutrons and thermally thick

The attenuation path length  $\beta_s$  increases when the enrichment of  $U^{235}$  in the uranium sample increases.

In Fig. 2 we can see that the fission of  $U^{235}$  nuclei located between 0 and  $-(L_s - l_s)$  does not contribute to the pyroelectrical signal. So, it is necessary to make two corrections. The first one, because the sample thickness that contributes to heat the pyroelectric detector is  $l_s$  and not  $L_s$ ; the second is due to the neutron flux. The uranium layer that is not contributing to heat the pyroelectric detector, attenuates the neutron flux. So, the neutron flux in  $-(L_s - l_s)$  is

$$\Phi'_o = \Phi_o \exp [ -\beta_s(L_s - l_s) ] \tag{11}$$

The sample-air boundary is replaced by the boundary in  $-(L_s - l_s)$ . Therefore:

$$b_{sg} = b_{ss} = 1 \tag{12}$$

By replacing (12) in (10), we can obtain:

$$V(\omega_o, \beta) = \frac{p \Phi'_o E}{2\epsilon} \left( \frac{\beta_s}{K_s(\beta_s^2 - \sigma_s^2)\sigma_p} \right) \left[ \frac{r_s + 1}{b_{ps} + 1} (e^{-\alpha_s l_s} - e^{-\beta_s l_s}) \right] \tag{13}$$

By making:

$$\frac{\beta_s}{K_s(\beta_s^2 - \sigma_s^2)} = \frac{\beta_s}{K_s \sigma_s^2 (r_s^2 - 1)} = \frac{\beta_s}{K_s \sigma_s^2 (r_s - 1)(r_s + 1)}, \tag{14}$$

we can simplify (13):

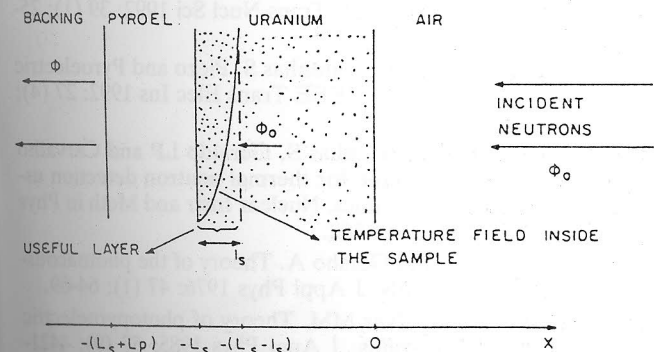


Fig. 2 - Neutrons self-absorption in the sample.

$$V(\omega_o, \beta) = \frac{p \Phi'_o E}{2\epsilon} \left[ \frac{\beta_s}{K_s \sigma_s^2 (r_s - 1)\sigma_p} \right] \times \frac{e^{-\alpha_s l_s} - e^{-\beta_s l_s}}{(b_{ps} + 1)} \tag{15}$$

The useful layer is thermally thick ( $\mu_s < l_s$  and  $\mu_s \ll \mu_{\beta s}$ ). Then  $\exp [-\alpha_s l_s] \cong 0$ .

If the thermal absorption is larger than the attenuation absorption (the sample has low enrichment and  $\beta_s$  is small),  $|r_s| \ll 1$ ,  $e^{-\alpha_s l_s} \ll e^{-\beta_s l_s}$  and  $b_{ps} \gg 1$ . We can obtain:

$$V(\omega_o, \beta) = \frac{p \Phi'_o E}{2\epsilon} \frac{\beta_s}{K_s \sigma_s^2 \sigma_p} \frac{e^{-\beta_s l_s}}{(b_{ps} + 1)} \tag{16}$$

By replacing (11) in (16) we obtain:

$$V(\omega_o, \beta) = \frac{p \Phi_o E}{2\epsilon} \frac{\beta_s}{K_s \sigma_s^2 \sigma_p} \frac{e^{-\beta_s l_s}}{(b_{ps} + 1)} \tag{17}$$

The frequency of  $V(\omega_o, \beta)$  in (17) varies as  $\omega^{-3/2}$ . When the sample has high enrichment  $|r_s| \gg 1$ ,  $\beta_s/\sigma_s \gg 1$ , we can obtain:

$$V(\omega_o, \beta) = \frac{p \Phi'_o E}{2\epsilon} \frac{e^{-\alpha_s l_s}}{K_s \sigma_s \sigma_p (b_{ps} + 1)} \tag{18}$$

The signal in (18) varies as  $\omega^{-1}$ .

3. Materials and methods

The detector system consists basically of a lead zirconate titanate (PZT<sub>4</sub>) ceramic (17 mm x 17 mm and 3 mm thick) from Edo Western Corporation, attached to an uranium ( $U_3O_8$ ) pellet with 14 mm diameter and 2 mm thickness. A good thermal coupling between ceramic and uranium has been obtained by using an Apiezon-L type grease. The neutron beam was modulated by a chopper built with cadmium foil 1 mm thickness. The irradiations have been carried out at a beam tube (BH-8) of the IEA-R1, 2 MW pool type research reactor. The thermal neutron flux has been measured by the activation technique with gold foils.

4. Results

The detector response for thermal neutron fluxes from  $8.0 \times 10^3$  to  $8.5 \times 10^5$  neutrons/(cm<sup>2</sup>.s) has been experimentally studied, at the chopping frequency of 3.0 Hz, and the results are shown in Fig. 3. Figure 4 shows the pyroelectrical signal as a function of the inverse of the chopping frequency. With a microcomputer it was possible to adjust the curve described by eq. (18) to the experimental data, with the deviation:

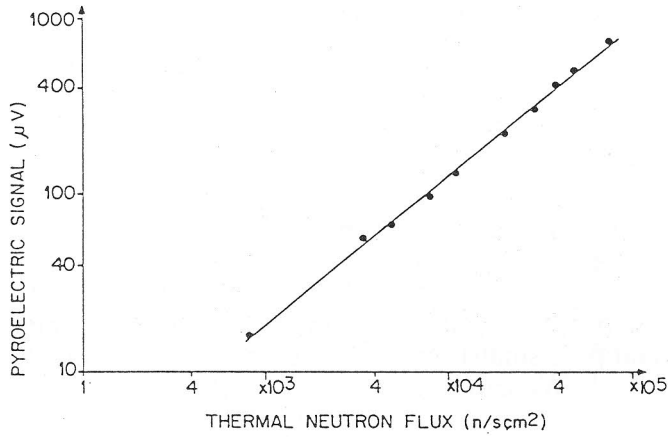


Fig. 3 - Plot of pyroelectrical signal vs thermal neutron fluxes, at the chopping frequency of 3.0 Hz.

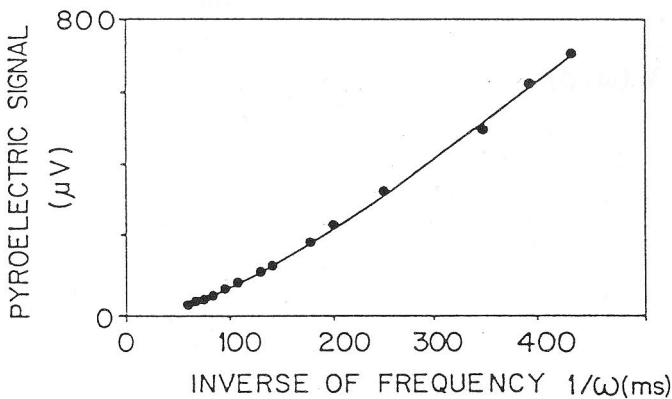


Fig. 4 - Pyroelectrical signal as a function of the inverse of the chopping frequency. Sample:  $U_3O_8$  pellet with 20% enrichment in  $U^{235}$ .

$$d = \frac{[\sum_j (M_{ji}I - Y_j)^2]^{-1/2}}{\text{Number of data (M)}} \quad (19)$$

where  $j$  represents the elements of a matrix that contains the frequency data ( $M$ ) and the pyroelectrical signal ( $Y$ ).

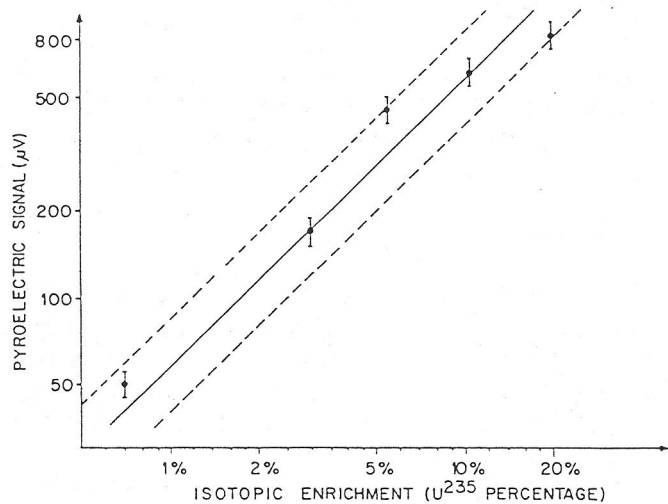


Fig. 5 - Plot of the pyroelectrical signal vs the uranium enrichment, at the chopping frequency of 3.3 Hz.

The pyroelectric signal as a function of the uranium enrichment, at a constant chopping frequency, is shown in Fig. 5. The pyroelectric signal was corrected because only the thickness  $l_s$  contributes to the pyroelectric signal generation and the sample thickness  $L_s - l_s$  only absorbs neutrons.

## 5. Conclusions

In the theoretical model presented, we have classified the pyroelectric detector as transparent for neutrons and thermally thick and the uranium sample as semi-transparent for neutrons and thermally thick. For samples with high enrichment in  $U^{235}$  the pyroelectric signal is inversely proportional to the chopping frequency  $\omega$ . For samples with low enrichment the signal varies as  $\omega^{-3/2}$ . As can be seen in Figs. 3, 4 and 5 the waveforms determined from the theoretical model agree well with the experimental data. By using the developed theory it is possible to determinate thermal parameters of nuclear fuels, for example the thermal diffusivity. It is possible also to determine if a sample has a low or a high enrichment in  $U^{235}$ .

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