Temperature Dependence in the Long-Term Stability of the TlBr Detector

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***Abstract*—In this paper, TlBr detectors with three-electrodes were prepared and their long term stability evaluated as a function of the temperature. Systematic measurements of counting rate were carried out to observe the random pulse formation, due to the polarization effect. Three-electrode detectors presented stability of about 112 h at near room temperature and more than 325 h at 0°C. Although the three-electrode detector presents a better performance compared to the two-electrode detector, its stability has been still dependent on the temperature.**

***Index Terms*—Detector stability, polarization, semiconductor detector, Thallium bromide.**

I. Introduction

THALLIUM bromide crystals present promising characteristics for use as room-temperature semiconductor detectors. TlBr has a band gap energy of 2.7 eV, high atomic number ($Z_{Tl} = 81$ and $Z_{Br} = 35$), high density (7.5 $\mathrm{g/cm^3}$), high resistivity ($10^{12}\,\Omega\mathrm{cm}$) and melting point at 480°C [1], [2]. However, TlBr detector use has been limited due to a polarization effect, possibly attributed to ionic conduction [3], [4], which reduces the long-term stability. It was observed in the literature efforts to investigate the polarization effects and long-term stability of the TlBr crystal detector [1], [5]–[7]. Onodera *et al.* [1] obtained stability of about 10 h at room temperature and 100 h for −20°C for TlBr detectors made with two electrodes, improving the crystal quality. Hitomi *et al.* [6] enhanced the stability using detectors with the small pixel technique. Hitomi *et al.* [5] improved the two-electrode detector stability by adding Thallium to the electrodes. More recently, the authors [7] obtained a full stable device, changing periodically the bias voltage polarity. In this paper, TlBr detectors were prepared with three-electrodes and characterized as a function of time, at different temperatures, by measuring the counting rate. The stability of the detector was dependent on the temperature. The three-electrode detector configuration used in this work was more stable compared to that obtained with the two-electrode detector used in our previous works [8], [9].

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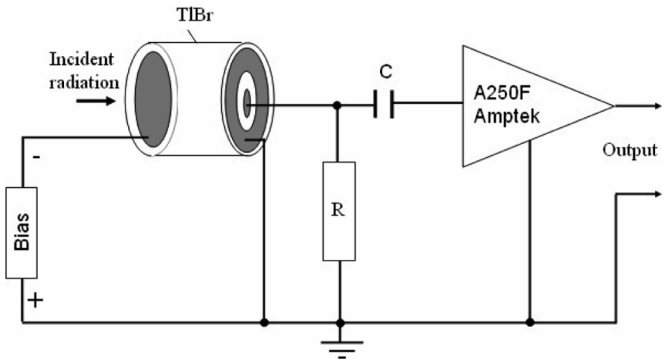

Fig. 1. TlBr detector and preamplifier connection.

II. Device Fabrication

TlBr crystal was grown by the vertical Bridgman technique, using a quartz crucible in vacuum. A crystal around 10 mm diameter and 20 mm long was obtained with a growth rate of 1 mm/h. The crystal was sliced in wafers, cut transversally to direction [110], using a diamond saw and lubricated with glycerine during the cut. Five slices of TlBr crystal were cut to be used as detectors.

After cutting, the TlBr slices surfaces were treated using the procedures that presented better results in our previous studies [8], [9]. In those works, better results were obtained for TlBr samples without chemical etching, after polishing, and with electrodes made using colloidal carbon in organic solvent from Viatronix ™. Therefore, this procedure was used to prepare the TlBr surface.

Among the five slices cut, two of them were polished, cleaned and the electrodes painted subsequently, without pause. For the other three slices, the procedure of the electrodes painting was performed around three hours after the polishing. The final thickness of the crystals was about 0.5 mm.

The three-electrode detectors were made with a central electrode (anode), plus a ring electrode surrounding the central electrode and the third electrode (cathode) at opposite crystal face. Fig. 1 shows a schematic diagram of the detector and its connection to the preamplifier. The central electrode diameter has approximately 3 mm, the ring electrode diameter was made as large as possible, around 9 mm, and the distance between these electrodes was around 1 to 2 mm.

III. Methods

Fig. 2 shows the measurement schematic diagram. The experiments were carried at four different temperatures, with the

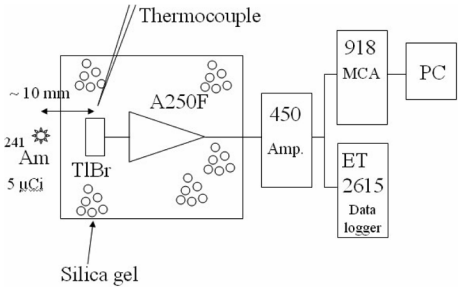

Fig. 2. Schematic diagram for the measurements of a TlBr detector at different temperatures.

detector set wrapped in a box with silica gel. For lower temperatures, the detector was placed in a refrigerator. The temperature was measured with a thermocouple near the detector. The silica gel was used to prevent the humidity in the detector inside the refrigeration system, because at low temperatures, the water condensation around the crystal was enough to suppress any signal from the detector. The output from A250F charge sensitive preamplifier was connected to 450 EG&G Ortec Research Amplifier, at 10 μs shaping time, and to EG&G 918A Multichannel Analyzer, to obtain the pulse height spectra. The counting rate was measured for different temperature ranges. The experiments were carried out starting at the warmest temperature, 28°C to 30°C. The pulse rate counting and its saving were carried out connecting the output of 450 EG&G Amplifier to a Brymen ET-2615 data logger, which was used as a ratemeter. An $^{241}\mathrm{Am}$ (60 keV) radioactive source was used for all measurements.

IV. Results and Discussion

Firstly, the detector was used as a DC coupled, but the bias voltage through the detector stopped the A250F preamplifier from working. This effect could be attributed to the low resistance of the detector, about $7 \times 10^9\ \Omega$ between the bias source and the preamplifier, changing the A250F electrical conditions. The resistivity value found for the detectors used in this work was about $10^{10}\ \Omega\mathrm{cm}$, lower than the listed value of $10^{12}\ \Omega\mathrm{cm}$, in the Table of [10]. To overcome this limitation, the detector was AC-coupled to apply higher bias voltage without compromising the A250F electrical conditions.

The TlBr slices were prepared with polishing, cleaning and electrode painting procedures, without pause. In these conditions, the maximum bias voltage reached around 300 V, however a few random pulses could be observed. Thus, the 200 V bias was used for all the experiment to assure no presence of this random current burst that produces undesirable pulses. The use of 200 V bias allowed the 60 keV photopeak from $^{241}\mathrm{Am}$ to be observed, as shown in Fig. 3.

For the TlBr slices prepared with a three-hour pause between the cleaning and electrode painting, the maximum bias voltage possible to apply was 150 V. To verify these results, the electrodes of these three detectors were removed and the TlBr slices were polished again, cleaned and the electrodes painted subsequently, but this time, without pause. Thus, the maximum bias voltage obtained for these three detectors was also about

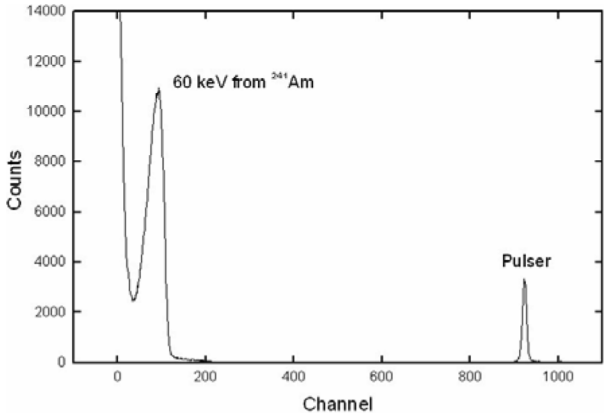

Fig. 3. Typical photopeak from $^{241}\mathrm{Am}$ for TlBr detector with three-electrodes and 200 V bias obtained during 300 s, at 17°C to 20°C temperature range.

TABLE I
Maximum Bias Voltage Versus Range Time Between the Crystal Polishing and Electrode Painting

	Maximal Bias Allowed (V)	
	Crystal#1	Crystal#2
0	300	200
30		140
90		100
180	150	

300 V, as obtained for those two first slices, indicating that the polishing, cleaning and electrode painting procedure should be made without pause.

All five samples presented similar pulse height spectra to that shown in Fig. 3. To confirm this behavior, contacts were painted at different time ranges using a second crystal. The obtained results for both crystals are summarized in Table I. From this result, to paint electrodes immediately after the polishing is recommended.

It should be mentioned that in the procedures of crystals painting removal, the ring electrodes showed a migration of the carbon colloidal painting into the crystal to the cathode surface direction. For removing the carbon colloidal from the ring electrode was necessary sanding and polishing the ring crystal surface. For the crystals used for about 500 h, it was necessary to remove about 0.05 mm of the thickness for removing the migrated carbon. However, for the central electrode, the painting was easily removed with alcohol or acetone, indicating low carbon migration. Despite the central and ring electrodes were at the same potential, the central electrode was connected to the ground using a high value resistor (200 $\mathrm{M}\Omega$). This high value could reduce surge currents in the central electrode due to polarization effects, avoiding the carbon migration by this electrode.

The long-term stability was evaluated with the TlBr detector working as a ratemeter. Figs. 4–7 show typical counting rate behavior, as a function of the temperature, for the sample studied. Similar results were obtained for all five samples prepared in this work. As it can be observed in Figs. 4–7, the counting rate

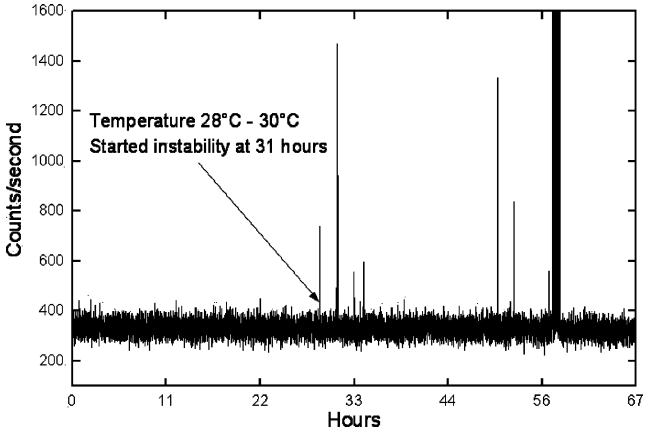

Fig. 4. Counting rate of TlBr detector vs. time at 28°C to 30°C range.

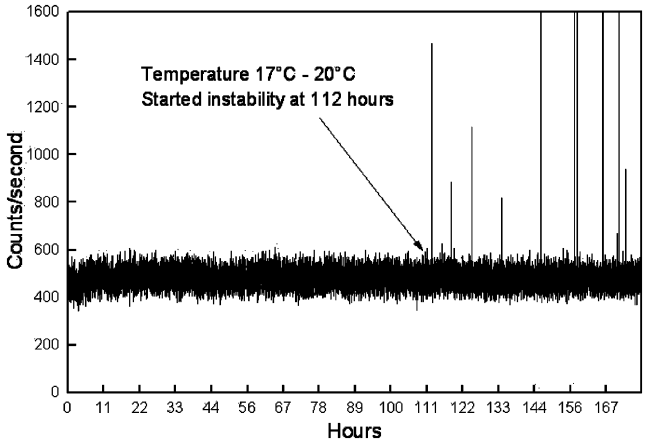

Fig. 5. Counting rate of TlBr detector vs. time at 17°C to 20°C range.

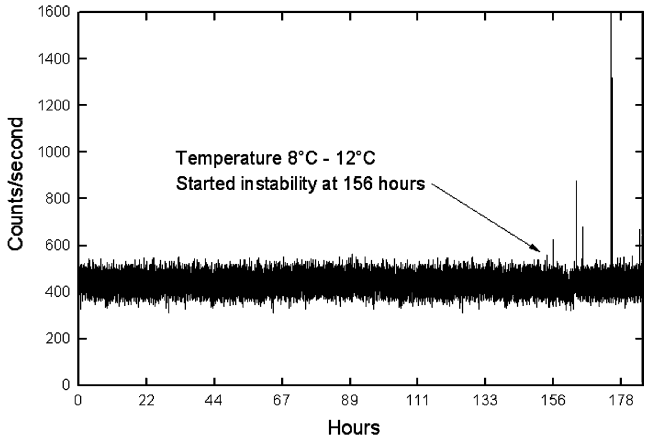

Fig. 6. Counting rate of TlBr detector vs. time at 8°C to 12°C range.

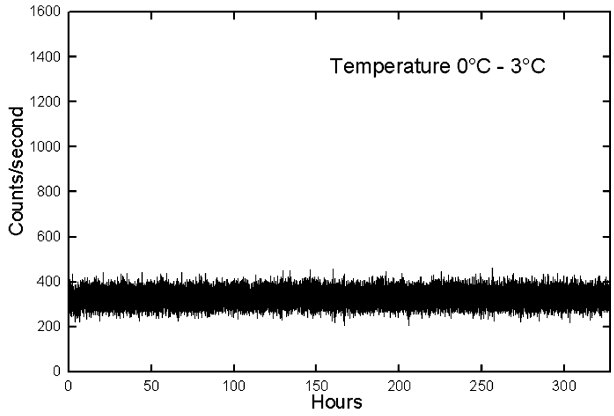

Fig. 7. Counting rate of TlBr detector vs. time at 0°C to 3°C range, under 325 h.

increases in some points due to the incidence of random pulses. Vaitkus *et al.* [3] had described this phenomenon as discharge of polarization and Dönmez *et al.* [11] as a breakdown. This phenomenon was not remarkably observed in the pulse height spectrum measurement, in our studies. This may be because the random pulse does not affect the energy spectra significantly, due to its random amplitude characteristics and low rate at the configuration used in this work.

Before starting the detector set-up as a ratemeter, at different temperature ranges, the bias voltage was removed for about 2 h, before each measurement. This recovery time was necessary so that the detector returns to the initial conditions, that is, without presenting random pulses. Fig. 4 shows the counting rate of TlBr detector as a function of the time at a temperature range of 28°C to 30°C. The instability from the polarization effect started at about 31 h of continuous bias application. Fig. 5 shows the counting rate at 17°C to 20°C range and the instability started after 112 h. Fig. 6 shows the counting rate at 8°C to 12°C range and instability starting after 156 h. Fig. 7 shows the counting rate at 0°C to 3°C range. In this last temperature range, the measurement was carried out over 325 h. During this period, instability in the detector was not observed. Further experiments should be performed to determine the real long-term stability at 0°C to 3°C range.

Comparing the present results with our previous work [8], the TlBr detectors made with three electrodes showed better stability than detectors made with only two electrodes (without the ring electrode). In both works, the detectors were made from the same crystal sample.

The detector stability was evaluated according to two models: linear and exponential as shown in Fig. 8.

The linear model presented a suitable fitting in the range from 10°C to 30°C. According to this model, for the temperature from 0°C to 3°C range, the breakdown is expected to occur in around 228 h, but this event was not observed in that time. On the other hand, the use of an exponential model (Fig. 8) could be considered more realistic although it is statistically more complex because the curve fitting needs to use the method of nonlinear least squares. In addition, the uncertainties in its parameters were considerable due to the imprecision in the x–y variables and the small number of experimental points available for this analysis ($n = 3$). The exponential model predicts the breakdown to happen in around 343 h. However, as the breakdown event could not be observed in this work because the experiment was carried out only for 325 h. Therefore, this result, although not planned, is in conflict with the linear model, but it is in agreement with the exponential model. On the other hand, as the main interest is to use the TlBr semiconductor detector

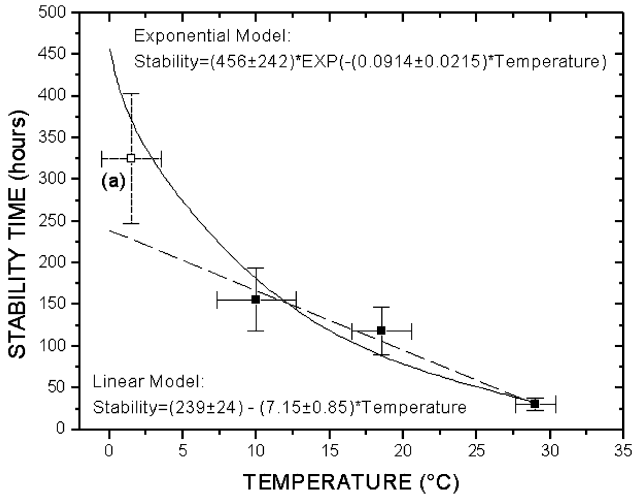

Fig. 8. TlBr stability (time to start the random pulses) versus temperature predictive models. The estimated error (1 standard deviation) in the stability was assumed as $\sim 24\%$ of the stability value and 68% of the respective temperature range (Table II). (a) For the point value of $0°\mathrm{C}$ to $3°\mathrm{C}$, the stability assay was carried out for 325 h but at this time the breakdown was not observed. This point is included in the figure with the purpose of comparing the capacity of prediction for each model but it was not used in the regression analysis to estimate the model parameters. For both models their parameters were estimated by considering only the last three points ($n = 3$) and the weighted least-squares regression were applied considering the errors in both variables (x and y).

at room temperature, both models, linear or exponential, can be considered satisfactory to predict the effect of the temperature on the stability of this detector. For the linear model, we can infer that the stability $\boldsymbol{S}$ of the detector decreases by 7.15 h for each increase of $1°\mathrm{C}$. Furthermore, for the exponential model, the stability is expected to degrade according to the function

$$\frac{d(S_0 \cdot e^{-\lambda \cdot T})}{dT} = -\lambda \cdot S_0 \cdot e^{-\lambda \cdot T} \frac{\text{hours}}{°C} \tag{1}$$

where $\lambda(°\mathrm{C}^{-1}) = 0.0914 \pm 0.0215$ and $S_0(\text{hour}) = 456 \pm 242$. Particularly, at $20°\mathrm{C}$ the stability $\boldsymbol{S}$ is expected to decreases 6.7 h per $1°\mathrm{C}$ and so on. Further studies should be performed to validate the model and to define its accuracy parameters, in order to predict the stability of the detector for a wider temperature range.

The instability due to polarization effect did not occur continuously for three-electrode detectors as occurred with the detectors made with just two electrodes, that is, with the ring disconnected from the ground. For two electrode-detectors, it was only possible to decrease the polarization effects by reducing the bias voltage or changing the bias polarity. However, for three-electrode detectors, just removing the bias for some hours was enough to return the detectors to the initial conditions.

Twenty days after the detector was kept without bias, new measurements at $17°\mathrm{C}$ to $20°\mathrm{C}$ range were performed to evaluate the reproducibility of the stability. The first new measurements presented unexpected values (18 h and 34 h to start the instability). These results were not expected since they were not

TABLE II
TLBR EXPERIMENTAL CHARACTERIZATION

Temp. (°C)	Stability (hours)[a]					Average (hours)
0-3	>325					>325
8-12	156					156
17-20	112	80[c]	156[c]	112[c]	130[c]	118.0 ±2 7.9 (24%)[b]
28-30	31					31

[a] Hours to start the random pulses
[b] Mean ± 1 standard deviation (percentage error)
[c] Measures performed 20 days after the detector was kept without bias

comparable with those obtained in the measurement shown in Fig. 5 and Table II.

It was verified that the humidity in the three-electrode detector, due to the air moisture presence in the detector box, degraded the detector stability. Therefore, to remove the moisture, silica gel was heated and placed again inside the detector box and the detector was biased. After this procedure, four other measurements (labeled with "c" in Table II) were carried out, with results considered satisfactory and values comparable to the 112 h presented in Fig. 5 and Table II. It should be emphasized that the random pulses occurred in a short time (18 and 34 h) due to the humidity present in the air. These pulses were not distinguishable from the random pulses produced by the polarization effect. These results suggest a dependence on the environmental condition (humidity) around the detector to produce random pulses. This fact also suggests that the instability of the detector can be understood as a combination of the polarization effect and the humidity. This last effect could be minimized when the detector is maintained in dry atmosphere, vacuum or properly sealed. In our previous works [2], [8], [9], [12], using two-electrode detectors, the silica gel use was not required because the humidity did not interrupt the functionality of two-electrode detectors at any temperature conditions. On the other hand, the two-electrode detector presented a progressive reduction of the charge collection, while the three-electrode detector did not show the same behavior. For three-electrode detectors, the expected decrease of charge collection with the increase of the trapped charges did not occur, due to the electrode-ring be connected directly to the bias and it worked as an efficient trapped charges scavenger. Another charge collection aspect observed was a progressive pulse height increase, as shown in Fig. 9.

For new three-electrode detectors, not yet submitted to the bias tension, an initial increase of the pulse height of 7.5 ± 0.3 channels/day was observed (Fig. 9). The plateau was reached in approximately 25 days. However, in the subsequent use of the detector, after a rest period, it did not present so much initial variation of the pulse height. In this case, in our experiments, the first measurement already presented a pulse height located around 10 channels below the plateau channel level (Fig. 9).

The results of the pulse height increase, shown in Fig. 9, can not be understood as a better charge collection due to a trapped charge reduction, but because of the effective crystal thickness reduction which increased the charge collection. This effect was observed in our crystals due to the poor mobility property. It

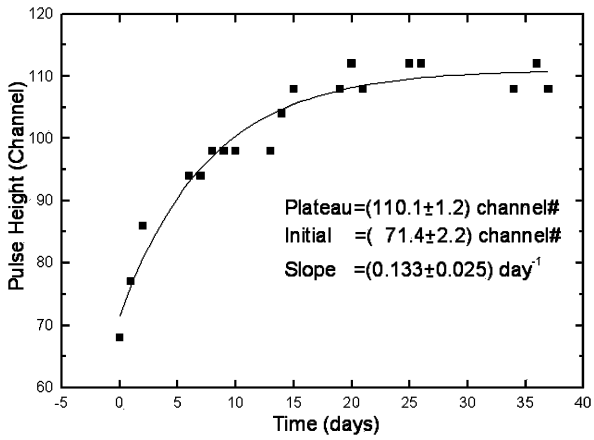

Fig. 9. TlBr detector pulse height time dependence using a $^{241}\mathrm{Am}$ (60 keV) source and 200 V bias.

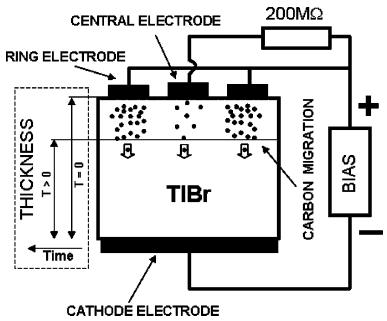

Fig. 10. Scheme of the carbon migration in the TlBr substrate and its effective (virtual) thickness for detectors made with electrodes painted with colloidal carbon.

can be overcame by reducing the crystal thickness. In an ideal crystal, for the same charges number produced, the number of charges collected in the electrodes is nearly independent on the crystal thickness. Thus, the pulse height should be equal for any thickness of the same crystal. On the other hand, defects or impurities in crystals can degrade the mobility or the charge collection. Therefore, due to these concentrations, the same crystals, with different thicknesses, can present different pulse height. Fig. 10 is a model to explain the results shown in Fig. 9. This model introduces the idea of the colloidal carbon migration in the TlBr crystal bulk for a detector prepared with three electrodes. The carbon migration was already observed by Vaitkus *et al.* [3] and characterized as a fractal type migration. The similar behavior for Tl ions also was observed by Hitomi *et al.* and [7], Vaitkus *et al.* [13]. The carbon migration as the time passes, induces a reduction of the virtual crystal thickness. As the used crystal presents defects and impurities in its structure, the reduction of the thickness provides a lower trap number in the pathway of charge movement and, consequently, an increase in the charge collection. Thus, the increase in the pulse height, shown in Fig. 9, can be explained by the virtual thickness reduction of the crystal, along the time, produced by the carbon diffusion in the TlBr substrate reducing the crystal trapping length [14]. Then, due to the evolution of the carbon migration, the total degradation of the detector is expected with the fall of its resistivity and loss of the semiconductor properties as radiation detector. Therefore, the plateau observed in Fig. 9 should not be kept indefinitely, and the pulse height can be reduced up to its annihilation. Further studies should be carried out to estimate the detector real life.

In all five samples studied in this work, when the ring electrode was grounded, the pulse rate increased and it was dependent on the ring area painted on the crystal. This can be attributed to movement of collected charge to the ring electrode, which induces charge in the anode electrode and increases the number of counts. This pulse rate dependence it is not expected for an electrode, working as a guard ring, thus, in this work it was called just ring electrode.

V. Conclusion

The higher bias voltage and better stability were found for the detectors polished, cleaned and their electrodes painted immediately after the polishing. The humidity present in the environment around the detector reduces its stability, thus it is recommended the detector should be maintained in dry atmosphere, vacuum or properly sealed. TlBr detectors prepared with three electrodes presented a better long term stability compared with two-electrode detectors. In spite of the increase in the stability using three-electrode detectors, the temperature is still a limiting factor for continuous operation. The stability of the detector can be predicted by the linear model in the range between $10^{\circ}\mathrm{C}$ to $30^{\circ}\mathrm{C}$ but the exponential model is more realistic for entire length of temperature. The continuous use of the detector increased the pulse height up to achieve a plateau. This fact can be explained by the effect of the carbon migration contained in the paint applied to the ring electrodes due to the virtual thickness reduction of the crystal as time passes. For new detectors, not yet submitted to the bias tension, the pulse height increased its level of 7.5 channels/day. The plateau was reached in approximately 25 days.

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