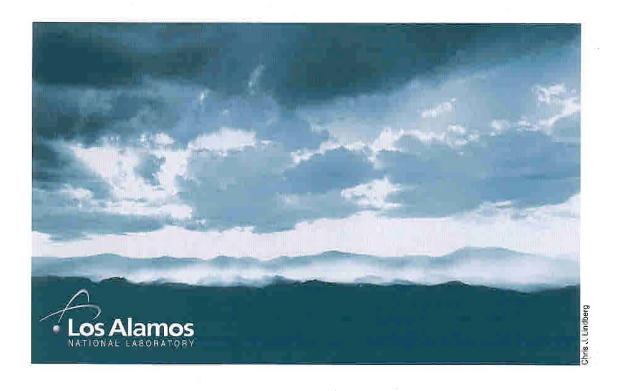
Temperature Behavior Of The Doped Nai (Tl) Scintillators And Its Impact On The Pulse Height Analysis Instrumentation

Boian Alexandrov, Jeff Audia, Michael C. Browne, Kiril Ianakiev, Ha Hguyen, and Pam Reass Los Alamos National Laboratory Los Alamos, NM 87544 NM

Presented at the
Institute of Nuclear Material Management
46th Annual Meeting
Phoenix, Arizona
July 10-14, 2005



This is a preprint of a paper intended for publication in a journal or proceedings. Because changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

Temperature compensation of NaI (Tl) scintillation detectors

Boian Alexandrov, Jeffrey M. Audia, Michael C. Browne, Kiril Ianakiev, Ha Hguyen, Pam Reass, Robert Parker Los Alamos National Laboratory 87544 NM

Abstract

The temperature dependence of inorganic scintillators is nonlinear over the temperature range for outdoor implementation, mainly due to the complex change of the light yield as well as light decay time over the temperature range. Our spectrum stabilization approach is based on rough and prompt control of effects of the temperature drift that keeps the reference peak within +/-5%, as well as software peak tracking for precise correction of the energy calibration and long-term aging effects. A peak deviation less than +/- 2% was measured for the temperature range -20°C to +60°C. The experimental results and temperature behaviour of the system based on 4"x16"x2" slab detectors will be discussed.

Temperature behavior of the doped NaI (Tl) scintillators and its impact on the pulse height analysis instrumentation

Boian Alexandrov, Jeffrey M. Audia, Michael C. Browne, Kiril Ianakiev, Ha Hguyen, Pam Reass, Robert Parker

> Los Alamos National Laboratory 87544 NM

1. Introduction

A Sodium Iodide detector doped with thallium activator, NaI (TI), is one of the most commonly used low-resolution room temperature gamma detectors because of its unbeatable efficiency per unit cost, especially in MeV energy range. The limitation in its implementation comes mostly due to its gain instability over the temperature and time, rather than its low resolution. The temperature dependence of the scintillator's light output is complex, as both the size and shape of the light pulse change with temperature. The basic properties of NaI (TI) scintillators have been well studied and understood by condensed matter scientists ^{1,2}, but some of the properties, such as multicomponent light output, have been forgotten over the years by detector manufacturers and users.

The basic understanding of NaI (Tl) detectors was simplified as a temperature-dependent single decay time constant ^{3,4} and represented by widely used Harshaw experimental data ⁵. The existence of two dominant components, with fast and slow time constants, in the output signal of the NaI (Tl) scintillation detectors was precisely observed long ago⁶, but in the same paper a conclusion was made that there is no need to take into account the slow component, except via appropriate temperature dependence of a single time constant. Even knowing this, the effect of the changing light decay time on the signal processing was not taken into account. Bicron show more linear data taken with a longer shaping time constant⁷, but does not explain its effect on the pulse height spectrum.

Our study of NaI (Tl) shows that the dependence of the total light yield is linear for the entire temperature range for outdoor application and two dominant decay time constants with temperature dependent redistribution of the intensities. The second decay component is negligible at room temperatures but it occupies up to 40% of the total light and duration of several microseconds at temperature -20C° ⁸. The effect of this complex and temperature dependent shape of the light pulse on the overall parameters of the NaI (Tl) spectrometer as a pulse height gain, deterioration of the resolution, dead time, is discussed in ⁹

2. Light pulse experimental data and modeling the effects of temperature

Our model for the temperature dependent behavior of the NaI (Tl) scintillators is based on two dominant components of the light pulse: one fast with a time constant of 230 nsec and one slow with a time constant of about 1µsec. These two components correspond to two different processes in the crystal. The physical model of the process briefly is:

In the very beginning we have production of the electron-hole pairs production along the path of the ionizating particle. Because of the very strong coupling between the electron and holes in alkali halide crystals, ¹⁰ Excitons are formed in the Exciton band followed by creation of the Self Trapped Excitons (STE) ¹¹. The fast component is the result of prompt capture of the Exitons by the (Tl^+) level ^{1, 2}.

We observe clear temperature redistribution of the amplitudes of the two different components, i.e. the redistribution of light output, which actually is the redistribution of the charges that belong to the different mechanisms. We believe this redistribution is via the temperature dependence of the population of the STE. The temperature dependence exists because the STE can reach the (Tl⁺) level via thermal assisted hopping and after that they can be captured and can recombine there. This process is competing with the multi-phonon-assisted dissociation of the STE. In this way, at every temperature there is a different part of the STE, which can "feed" the population of the Excitons responsible for the fast component of the light output. The rest of the STE, via multi-phonon-assisted dissociation, separates to the electrons and holes. After this multi-phonon-assisted dissociation, the separated holes via binary diffusion can reach the captured into the (Tl⁰) centers electrons and can recombine with them¹³. This second, mechanism is responsible for the amplitude of the second slower component of the light output of the NaI (Tl) scintillators. There is also a very weak temperature dependence of the time constants of the two processes because of the temperature dependence of the diffusion time constant.

Experimental data has been taken with a 2"x 2" Bicron detector MOD 3M3/3P, S/N DL-950, placed in an environmental chamber at temperatures ranging from -27C° to +50C°. An Ortec 113 preamplifier and six different shapers at constant room temperature outside the chamber were used. The temperature was ramped in steps of 15C° and held for 4 hours between steps to achieve temperature equilibrium in the entire volume in the crystal. The PMT's anode current as well as the pulse peak positions of 662 keV ¹³⁷Cs line for each shaper were recorded. Figure 1 shows the normalized areas of the current pulses at various temperatures. This figure shows the relative redistribution between fast and slow light components.

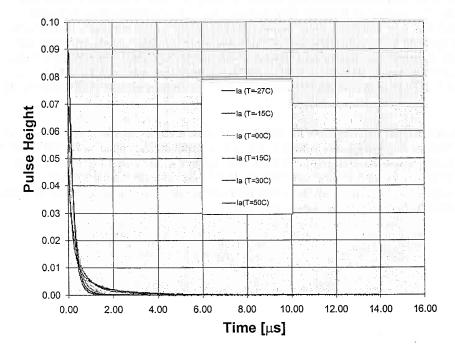
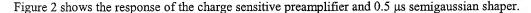


Fig. 1 Normalized to the total area of PMT anode current pulses for different ambient temperatures. The temperature dependent redistribution between fast and slow component is obvious.



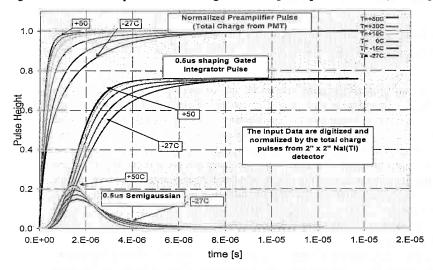


Fig. 2 Simulated response of the Preamplifier (upper), Gated integrator (middle) and 0.5 us Shaper (lower) to the normalized current pulses

The rise time of the preamplifier output pulse (equivalent to the total light) follows the duration of the PMT current pulse. The time for collecting 99% of the total light varies from 2µs at + 50°C to 10µs at - 27°C. The corresponding pulse height of the shaper with 0.5µs unipolar Gaussian pulse decreases monotonous with lowering temperature. Note that the lower the temperature, the lower the pulse height and the longer the pulse tail. This effect is well known as "ballistic deficiency", but the rise time variation is caused by the light decay, not by the carrier's collection time. It is well known fact (here would be a good place for a reference)that a shaper with a flat top in the weighting function equal to the preamplifier rise time variation could eliminate the ballistic deficiency. The middle traces of Figure 2 shows that a gated integrator response with integration time equal to the total pulse duration demonstrates this effect. Figure 3 shows the normalized ¹³⁷Cs peak position at +50°C taken with different shapers. When the slow component of the light output is taken into account, a linear temperature dependence of the peak position with negative slope is obtained. This negative slope is due to the temperature dependence of the integral light output due to the nonradiative phonon assisted transitions from the (T1+) levels to the ground level.

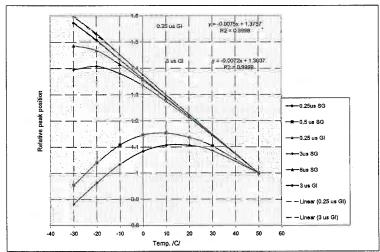


Fig. 3 Normalized to +50C 662keV peak position versus ambient temperature from different shapers. These data demonstrate the profound effect of the shaping on the temperature behavior of scintillators pulse height spectrum.

The gated integrator shaping shows excellent linear temperature dependence over the entire temperature range of -30°C to +50°C independent of the semigaussian prefilter time constant. The constant slope of the temperature drift is caused by phonons assisted nonradiative transitions as mentioned above in addition to the negative gain temperature coefficient of the PMT.

3. Our concept for temperature stabilization – a prompt temperature compensation and precise peak tracking

Most of classical methods for temperature and gain stabilization rely on the use of reference peak in the spectrum. The peak is generated by a gamma or alpha radiation source or an LED pulse source. In the case of an alpha or LED light source, the difference between the temperature dependence of the alphas or LED-induced light and that of gammas needs to be taken into account. Use of a reference gamma source allows for the direct tracking of the gain but it contaminates the pulse height spectrum. The activity of the reference source depends on the dynamics of the crystal's temperature and the background under the peak. Usually the tracer isotope is selected so that the gamma energy is high enough such that the statistical fluctuations of the reference peak do not deteriorate the intrinsic resolution of NaI (Tl) detector. This becomes a real challenge for small handheld crystals with a low peak to Compton ratio and fast-changing temperatures.

Additionally, the classical peak tracking needs to accommodate sudden changes in the spectrum in the vicinity of the reference peak when other gamma sources appear.

The gain shift due to temperature is relatively fast and could change the gain by a factor of two over the entire temperature range. Aging effects (mostly due to PMT gain) are very slow and accumulate observable drift over many weeks.

Our temperature stabilization procedure is based on two independent processes and takes into account the above-mentioned two considerations. We have linearized the light response over the entire temperature range, allowing for prompt, simple gain compensation based solely on the NaI crystal temperature. The temperature compensates for both the crystal light output and the PMT temperature drift. The purpose of this compensation is to keep the temperature drift to within 3-5% over the entire temperature range and prevent sudden and rough changes of the spectrum. This process allows for precise tracking of low activity reference peaks, such as the naturally occurring ⁴⁰K peak, for long term precise stabilization. Our experiments with a Bicron 4"x2"x16" NaI detector show count rates of about 33 cps at 1460 keV due to background ⁴⁰K. When the natural background is not enough, a bag of KCl is used to boost the background. This approach allows precise tracking at minimal contamination of the spectra. The tracking system is disabled whenever counts within the reference peak ROI rise above the statistical limit, and in this event, only the temperature of the NaI crystal is used to compensate for gain drift.

Figure 4 shows that application of temperature-only compensation keeps the gain drift below $\pm -2\%$ range of average value of the peak position at temperature $25C^{\circ}$.

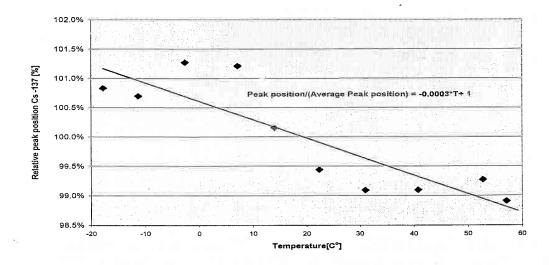


Fig. 4 Cs-137 peak position versus temperature, with temperature correction. The detector is Bicron detector 16''x4''x2'', S/N 70003-0025. The slope -0,03% is due to the temperature drift of the electronics

The SCIONIX TYPE - V102A102/3M-E1-X, 4"x4"x4" NaI (Tl) detectors prepared for installation in Chernobyl show very good consistency, thus general characterization could be used.

Detector	#7574/0001	#7574/0002	#7574/0003	#7574/0004
Temp drift [%/C°]	-0.2268	-0.2436	-0.2374	-0.2416

The average temp drift is -0.237 [%/C°] with STDEV 0.0075

4. The NaI (Tl) spectrometer configuration

Figure 5 shows the block diagram of the spectrometer used. The temperature sensor is attached by thermal conductive epoxy at the solid flange between the crystal and PMT optical interface. The gain compensation is performed in the preamplifier hardware based on the measured detector temperature. The signal processing consists of a 1µs unipolar Gaussian shaper followed by the gated integrator. The integration time of 8 µs was chosen as a compromise between compensation accuracy and throughput.

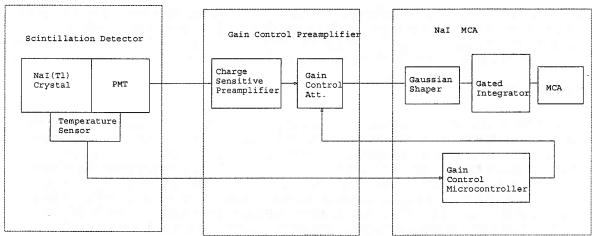


Fig. 5. Block diagram of NaI (TL) spectrometer with temperature compensation and rise time correction of the preamplifier pulse.

By mounting a temperature sensor external to the crystal and characterizing the thermal time constant, we can perform the same temperature compensation on off-the-shelf crystals. The inside temperature of the crystal differs from that of the detector's surface, thus causing a dynamic temperature-compensation error. Figure 6 shows the equivalent thermal time constant measured by the difference of the surface temperature response and transience of a ¹³⁷Cs peak position. In this instance, nearly two hours elapse before thermal equilibrium is reached.

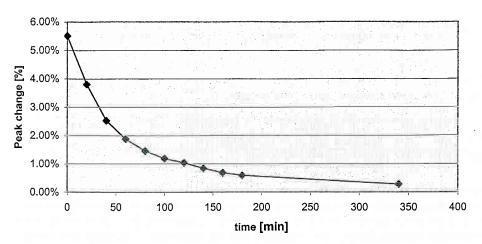


Fig. 6. Peak position of the Cs-137 versus time, after $10C^{\circ}$ temperature jump. The time constant is 1.9 hours.

In order to minimize this effect, thermal insulation of the crystal is required. This is easy in the case of portal monitors, where enough space can be provided for thermal shielding. For general purpose applications, an equivalent crystal temperature is calculated based on the measured step response. That virtual temperature takes into account the temperature delay by the thermal resistance of the insulation and thermal mass of the detector.

4. Conclusions

Temperature deviation in NaI crystals causes very complex changes in both total light output as well as the shape of the light pulse. The temperature dependence of the NaI (Tl) pulse height spectrum is a product of total light yield and convolution of the light pulse distribution and shapers pulse response. We have proven experimentally that total light output is linear over typical outdoor temperature applications. Use of gated integrator linearizes the temperature dependence, thus enabling simple compensation to be used. Further stabilization by reference peak tracking is necessary for long term gain shifts . Several 16"x4"x2" NaI(Tl) detectors and developed hardware have been characterized over the temperature range -20C° +60C°. The system detector/electronics with temperature compensation have been characterized, with a 662 keV peak deviation to within 2% for the range -20 C° +60 C°.

- 1. J. Birks, The theory and practice of scintillation counting, Pergamon Press, 1964.
- 2. R.Gwin and R.B. Murray Phys. Rev. vol. 131 (1963)
- 3. J.S. Schweitzer et al IEEE Transaction on Nuclear Science, Vol. NS-30, No. 1983
- 4. C.L. Melcher IEEE Transaction on Nuclear Science, Vol. 35, No1, (1988)
- 5. Harshaw catalogue
- 6. L. M. Bollinger and G. E. Thomas The Review of Scientific Instruments Vol. 32, (1961)
- 7. Rozsa, C. Grodsinsky, C. Penn, D. Raby, P. Schreiner, R. Bicron, Newbury Nuclear Science Symposium, 1999. Conference Record. 1999 IEEE.
- 8. K.Ianakiev, M.C.Browne, J. Audia, W. Hsue. Patent application S102, 322
- 9. K. Ianakiev et al, accepted in Nuclear Science Symposium, 2005 IEEE 2005
- 10. Van Sciever W.J., 1955 High Energy Physics Rpt, No 38 Stanford University (1955)
- 11. H. Nishimura and S. Nagata Journal of Luminescence 40&41 1988, 429-430
- 12. B. Goodman and O.S. Oen J. Phys. Chem. Solids Vol. 8, p. 291 –294 (1959)
- 13. R. B. Murray IEEE Transaction on Nuclear Science, Vol. NS-22, (1975)