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An Improved Method for the Stabilization of NaI-Photomultiplier Gamma Detectors Against Thermal and Other Drift

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Alpha peaks have been used as part of servo systems to stabilize NaI-photomultiplier gamma detectors against drift. However, alpha peaks shift with temperature change differently than do gamma peaks, thus spoiling what would otherwise be a workable scheme for stabilizing against probably the most serious source of NaI-p.m. detector drift, namely thermal effects. It has been found possible to accurately compensate for the difference in the shift with temperature versus gamma peaks using the signal derived from a thermistor in thermal contact with the NaI crystal to control the bias of a discriminator in the servo circuit. The servo circuit utilizing this principle has been used in commercial multichannel analyzers of the type intended for field use under adverse ambient conditions.

1. Introduction

Despite the more recent development of other detectors having specific advantages, the NaI-photomultiplier remains perhaps the most generally applicable detector for gamma photons, representing a useful compromise among the qualities of resolution, noise immunity, ruggedness, and portability.

A problem with NaI-photomultiplier based gamma detection has been gain drifts, i.e., short or long term changes in amplitude of pulses contributing to the full energy peak. There are several known causes for such gain shifts: Photomultiplier tubes are subject to hysteresis, fatigue, and several

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temperature induced effects, and the light output of scintillators for a given deposition of energy also changes with temperature.

Full energy peak stabilization schemes, either analogue or digital, which are based soley on the gamma spectrum being studied (as opposed to an external reference), have proved useful in laboratory applications of a specialized kind, e.g., the same photopeaks superimposed on similar Compton backgrounds, but are not as useful in portable or field applications, or where various mixtures of nuclides and or backgrounds are involved, and source intensities can vary over several orders of magnitude, and measurement geometries are not routine.

In portable or field applications, because of the latter circumstances, compactness, low power consumption, and simplicity of operation become paramount virtues. This seems to give preference to analogue control schemes based on an artificially introduced constant reference.

An analogue based stabilization scheme must necessarily involve as many parts of the detector-electronics system as possible, and in the same way as the pulses being stabilized. Thus, to stabilize gamma full energy peaks, it would seem appropriate to use a gamma ray from a long lived nuclide as a reference. The introduction of an extraneous gamma source to act this way however has faults: It is desirable that the energy of the gamma peak used for stabilization be appreciably higher than the energy of the gammas being stabilized from the standpoint of the overall servo gain. However, the Comptons from the stabilizing peak will then underlie the rest of the spectrum, contributing an unwanted background. This will not be negligible, since the reference count rate, needed to keep statistical fluctuations in the reference small, will turn out to be of the order of 10^3 1/s. On the other hand, if the peak used for stabilization is lower on the energy scale than the gammas of interest, not only is that bad from the servo gain standpoint, but also the stabilization peak will then be superimposed on a background of Compton pulses which will in general change shape from one assay or application to the next, thus effectively shifting the peak position.

An analogue stabilization scheme which does provide some relief in general applications and in field service makes use of a ^{241}Am alpha source exposed to the NaI crystal. This source produces pulses of constant gamma equivalent energy which are used to servo control the photomultiplier tube high voltage, hence the overall system gain.

The use of an alpha peak as a stabilization reference removes the particular difficulties mentioned above caused by using a gamma peak. Though alpha peaks will have an amplitude when generated in NaI approximately 0.5-0.6 of that of a gamma of equivalent energy, a typical alpha energy, say ~5.5 MeV, can NaI produce a pulse amplitude on the gamma energy scale of ~3 MeV, which is comfortably higher than the energies of many technically important gamma photons. There is moreover nothing comparable to a gamma ray Compton shelf in the case of the alpha pulse height spectrum; with reasonable care, the number of defective alpha pulses contributing to lower energy tailing will be negligible. ^{252}Am, half-life 432 years, which decays to ^{237}Np by alpha emission (~85% 5.48 MeV, ~13% 5.44 MeV) has proved especially useful.[1] So called "americium stabilized" NaI-photomultiplier detectors with appropriate electronics have been employed with some success for portable or field use.[2]

2. Ionization Density Effects

However, a stabilization scheme based solely on an alpha emitter neglects the dependence on ionization density of the change in pulse amplitude with temperature. This causes the alpha pulse amplitude, to which the servo is referenced, to change differently with temperature than the gamma pulse amplitudes being stabilized. This results in imperfect correction for the latter as regards temperature drift, though the servo system may still perform satisfactorily for the other sources of drift, such as fatigue, HV bias changes, etc.

We are unaware of any specific reference in the literature to this dependence on ionization density occurring in NaI, but suppose it to have elements at least qualitatively similar to known phenomena in certain scintillators which are in fact exploited in pulse shape discrimination schemes for separating neutron (recoil proton) and gamma (electron) caused pulses.[3]

The effect, illustrated below with experimental results, can in principle be explained assuming these hypotheses: (i) There are at least two mechanisms ("types of light emitting centers") from which light is emitted from NaI excited by ionizing radiation. (ii) The different mechanisms have different decay times. (iii) The relative proportions of types of centers available for light emission depends on the density of ionization (much greater for alpha particles than electrons). This could for example be brought about by non-radiative recombination "quenching" depending upon the local ionization density in different ways for the respective centers. (iv) The rates of non-radiative recombination for the different centers are differently affected by changes in temperature.

Such mechanisms pertain in specific instances for various organic and inorganic phosphors or scintillators and so it is reasonable to suspect similar for NaI(Tℓ). Particularly relevant are CsI(Tℓ) and CsI(Na) where pulse shape discrimination between charged baryons and electrons is possible.[4]

This effect of ionization density was studied in a superficial phenomenological way in connection with the development of a portable detector and pulse height analysis system for nuclear safeguards applications. These instruments[5] were routinely expected to be operable under extremes of temperature and yet required i.e., to yield precision (~0.1%) isotopic analysis based on relative gamma peak area. This was not possible without stabilization against thermal or other drifting.

3. Compensating for the Ionization Density Effect

After preliminary experimentation with other bases for stabilization such as gamma emitting isotopes or light emitting diodes,[6] we returned to the use of ^{241}Am alpha as a reference for reasons mentioned above, only to find it deficient as regards temperature drift. The solution to the problem turned out to be simple in principle and practice. Basically, it involved placing a thermistor in good thermal contact with the NaI crystal and using the temperature dependent signal from the thermistor to form a correction to the amplitude of the ^{241}Am alpha peak compensating for the peaks shift with temperature.

This correction is made simply with analogue circuitry with enough accuracy that there is no discernable (less than 1 part on 1000 peak shift) drift in the position of a full energy peak, over a range 5°C to 40°C, the permissable temperature limits for the detector. Without the thermistor, the drift over the same temperature range was greater than two FWHM (full width at half maximum).

In the following the components of the system will be described, followed by experimental data relating to the drift of peaks with temperature and the performance of the servo system.

4. NaI-p.m. Tubes with Alpha Sources and Thermistors

The method of making ^{241}Am - stabilized NaI - p.m. tube detector pioneered by Harshaw, uses a small piece of NaI crystal doped with ^{241}Am inserted into the main NaI(Tℓ) crystal so that it is in optical contact. Scintillations produced by the alphas in this "isotopic alpha light source" can thus be viewed by the photomultiplier through the main crystal. The Bicron process makes use of an external planchette ^{241}Am source placed in contact with the surface of a small piece otherwise ordinary NaI(Tℓ) crystal, this combination being inserted, or placed in contact with, the main crystal.

For either method there are three parameters to be considered in connection with a stabilization scheme that are introduced by the use of the ^{241}Am source: (i) The count rate is determined by the source activity. For the Harshaw type with a 4π geometry for the alpha source the count rate is the same as the activity, while for the Bicron type, with ~2π solid angle, it is about half. A useful count rate has been found to be about 1000 per second, which the manufacturer can control to about 10%. The statistical fluctuation in this count rate over the time period of the stabilizer circuit servo is satisfactorily small. (ii) Alpha particles are appreciably less efficient than gammas per unit energy in producing light. One can define the "alpha gamma-equivalent energy" (AGEE) of the alpha particles as the gamma ray energy that would produce the same (full energy peak) pulse amplitude as the particular alpha energy. The AGEE can be adjusted by varying the distance of the alpha light source from the photocathode. The maximum AGEE of ~3.5 MeV results if the light source is against the photcathode, while it might be <2 MeV for a light source ~10 cm away. As mentioned above, it is desirable that the AGEE be higher than the gammas of interest, but on the other hand it can be too high from the standpoint of overloading the subsequent amplifier stages, or forcing the spectrum of interesting gammas to be too tightly compressed within the amplifier's dynamic range. In the nuclear safeguards field, an AGEE of ~3 MeV has been found convenient since the highest energy gamma of common interest is that of ^{208}Tℓ at 2.61 MeV. The tolerance of AGEE obtained by the manufacturer is ~5%. (iii) The resolution of the alpha peak is of importance since it impacts directly on the gain of the servo system employing it. Alpha peak resolutions between 3 and 4.5% can be achieved by the manufacturer presumably by good optical and source of preparation technique. There is a certain inevitable amount of "tailing" of the alpha pulses, i.e., a lower amplitude distribution of "defectives" but this is small enough to be unimportant.

An interesting difference between the Harshaw and Bicron detectors: The 59.57 keV gamma emitted in prompt cascade with the 5.480 MeV alpha (~85% of decays) will not be seen with Harshaw type detectors since with the 4π light source geometry, it is simply summed into a total energy peak. With the Bicron type ~2π geometry, there is a finite probability that the gamma will be detected while the alpha particle preceding it isn't (or vice versa). In this case then the 59.57 keV line will show up as a separate peak in the gamma spectrum, where it can be used as a calibration point.

The earliest "Am stabilized" detectors were inferior in resolution, the reason being given that the insertion of the seed spoiled the light collection of the crystal. Recent Am stabilized detectors however have resolutions approaching the state of the art for ordinary NaI p.m. detectors, for example, ~7% for 5 cm x 5 cm (Harshaw), ~7.5% for 1.8 cm x 1.8 cm (Bicron), etc.

The stabilized detectors with alpha source and thermistor are distinctly more expensive than ordinary NaI p.m. tube detectors, costing in the neighborhood of $2000 for a 5 cm x 5 cm NaI p.m. detector. However, in addition to the ^{241}Am source and thermistor, in keeping with their probable use in a hostile environment, the p.m. tube comes with a prewired potted bleeder resistor string.

5. The Stabilizer Circuitry

The gain stabilization circuit taking advantage of the alpha source and thermistor is shown in block diagram form in Figure 1. Alpha particles from the source produce pulses whose amplitude corresponds approximated to that of a 3 MeV gamma full energy pulse. This alpha pulse is processed in the same amplifier used for gamma pulses except that it is taken out at a point before it saturates. It is sent to a discriminator whose bias is set relative to the alpha peak position so that half the alpha pulses exceed it. This bias, chosen for convenience to be 1 volt per MeV, depends on what the gamma equivalent energy is for the particular detector. Since the AGEE may differ even among detectors of nominally the same design, an adjustment is be provided. The thermistor to compensate for peak shift with temperature is in good thermal contact with the NaI crystal. This will be discussed in more detail below. Standard pulses from the discriminator are converted into a current proportional to the rate at which they are generated by an integrator circuit. This current, proportional to the count rate of alphas, is sent to a comparator whose other input is a current adjusted to be that appropriate to the nominal alpha count rate (ACR) of the source. The difference constitutes an error signal presumed to indicate deviation of the system gain from its desired operating point. The error signal is used to accordingly adjust the amplitude of an oscillator energizing the primary of a step up transformer whose rectified filtered output furnishes the high voltage (HV) for the detector bleeder string, therefore changing the p.m. tube gain.

Once the nominal settings for the alpha energy and count rate have been made for the particular detector in the laboratory or factory (AGEE and ACR controls), it is only required to set the HV adjustment to its nominal value. This is done with the aid of two LED indicators (see below). These indicate either the HV adjustment is too high or too low. Splitting the difference in the (10-turn helipot) setting gives the best nominal HV value. Once this adjustment has been made, the capture range of the feed back circuit is such (typically ±200V for a p.m. tube operating at ~700V) that the circuit will automatically stabilize at power up, i.e., no further adjustment of the HV need be made as long as the same detector is used with the system.

The fact that the control pulses are taken out of the amplifier before the last stage means that stage is not included in the overall servo gain control. This is however a negligible failing, as the gain stability of modern highly fed back solid state amplifiers is so good that they contribute negligible drift in comparison with a NaI p.m. detector.

The stability of the servo is such that it has made practical having several permanently precalibrated single channel analyzers (their biases derived from the same stable voltage source used by the stabilizer) adjusted to particular gamma lines of interest. Thus the instrument using this system is immediately ready for use without need for calibration or adjustment.

In the following, the circuitry making use of the thermistor and ^{241}Am source is described in more detail.

The alpha pulse signal from the p.m. tube passes through a conventional emitter follower acting as preamplifier and line driver (not shown). It then goes through the first stage (Figure 2) of the same two stage amplifier used for the gamma pulses. The first stage offers low noise amplification at low power consumption; all the transistors save one (2N3906) are contained on two CA3083 chips. The second stage (not shown) is similar. It is however designed to overload gracefully to accommodate the alpha pulse, which in the second stage will exceed the linear range (~4.5V), and also shapes the gamma pulses to a differentiated Gaussian.

The alpha pulse signal from C is then sent to the main body of the servo which is illustrated schematically in Figure 3. The LM219 serves as a distriminator whose bias is established by a 5 k pot. (the AGEE control) in series with the thermistor shunted by a ~1200 ohrm resistor; point Z is a regulated voltage.

The uniform amplitude pulses from the LM219 go to a diode pump arrangement, furnishing a smoothed output current derived from point Z via the 100 k pot. (the ACR control) and 10 M resistor. The output of the current comparator LH0042CD feeds into a long tailed pair which either transmits the alpha derived error signal to the high voltage (HV) regulating section or disconnects that error signal from the HV regulator, depending on whether the output of the LM219 discriminator is ungrounded ("stabilizer on") or not. The current comparator also incidentally indicates via two LED lamps whether the servo is in its control range.

The HV section can operate either as a stand alone adjustable (50 k, 10 turn, H.V. adjust pot) regulated bias supply, or as part of the servo system. The HV is generated with a commercial potted modular unit ("HV INV.") containing a multivibrator, oscillator, transformer, and high voltage rectifier. The amplitude of the oscillator, hence output voltage is determined by a discrete transistor (2N2905) which receives its signal from the LH004CD amplifier. The error signal to the LH004CD is derived from two sources, the bleeder string of 20 M resistors connected to the HV output, and from the alpha pulse derived signal.

The time constant of the HV regulator is ~100s. This is short compared to the time for any drift perturbation to occur, yet long enough so that the statistical uncertainty in the number of alpha pulses exceeding the AGEE discriminator threshold is small (<0.5%).

The capture range of the servo is fairly broad; typically if the HV is within ~±20% of its central, "best", value, the servo will lock in, as it will if the circuit was just turned on, if the HV adjust pot. is within the capture band.

Figure 4 is a realization of the regulator (except for the amplifier) on a single printed circuit card. This depicts a later version than that in Figure 3; note for example the HV polarity is reversible via a circuit board plug.

An important point, not realized until the circuit began to be adapted for use in other instruments, is the role of the amplifier time constant in the stabilization scheme. In retrospect this might have been expected in analogy to the pulse shape discrimination ability of certain organic scintillants. In that application, it is the difference in rise time of electron compared to recoil proton caused pulses that is exploited. The original development was in connection with an instrument which used a single differentiated Gaussian as a simple way to avoid count rate caused base line shifts. In adapting the instrument to a multichannel analyzer which was to be used with high resolution (Ge diode) as well as NaI p.m. detectors, it was desirable to leave the pulse Gaussian in order to optimise the resolution, where upon it was found necessary to increase the coupling capacitor at the output of the preamplifier from .001 to .01 μfd in order for the stabilization to function.

6. Performance of the System; Experimental Data

The action of the thermistor in compensating for the different shift with temperature change of alpha and gamma peaks may be best illustrated with data taken using the servo unit functioning properly as compared with the identical electronics disabled in particular ways. The temperature range of the measurements 5°C to 40°C corresponds approximately to the extremes.

Figure 5 shows the behavior of the peak positions for ^{241}Am and ^{137}Cs as the temperature of the detector is changed. Both peaks were displayed simultaneously on a multichannel pulse height analyzer (PHA) whose energy scale was established using gamma check sources when the detector was at 40°C. The resolution of the detector for 662 keV gamma rays is as shown on the same plot.

In this experiment, the thermistor was disabled in each of two runs, one with the ^{241}Am stabilization servo off (crosses) the other with it on (solid dots). From the "servo off" data, it can be seen that over the range 5°C-40°C the ^{241}Am peak which supposedly intended as a stabilization reference actually drifts more with temperature than the gamma peak. Quantitatively, the alpha peak shifts about 300 keV relative to a nominal value at 40°C of 2.7 MeV, or about -11%, compared to the gamma peak shift over the same temperature range of about 30 keV relative to the nominal value of 662 keV at 40°C, a change of -4.5%.

With the stabilizer on (the thermistor still disabled), the ^{241}Am alpha peak is sensibly constant over the whole range. The ^{137}Cs peak position which is actually the one of interest, now has approximately the same magnitude of drift, though in the opposite direction. What has happened in that the servo has functioned correctly in stabilizing the ^{241}Am peak, but since this peak shifts more with temperature than the ^{137}Cs gamma peak, the net effect of the servo on the ^{137}Cs peak is over compensation.

The working of the electronics with both the thermistor and ^{241}Am alpha peak stabilization operational is illustrated in Figure 6, which also shows the respective resolution curves for ^{137}Cs gammas and ^{241}Am alphas at 10°C. With the servo fully operational, the stabilization is being made against the position of the alpha peak as modified by the thermistor output.

As can be seen, the ^{137}Cs peak is now sensibly constant over the whole range 5°C to 40°C, but the ^{241}Am peak now shifts; the gamma peak is being kept constant because the thermistor change is accurately compensating for the shift in the alpha peak.

The experimental technique originally used to select the optimum value for the thermistor is illustrated in Figure 7. The basic thermistor value is nominally 1000 ohms with a 10% tolerance, deliberately chosen with a temperature coefficient known to be too great, i.e., such that it would over compensate. This was then shunted with an ordinary resistor and the relative shift in peak position divided by the temperature change plotted versus the value of the shunt resistance used. The resulting data is accurately represented by a least squares fitted line. The intercept on the R_S (thermistor shunt) axis, 1225 ohms, has the significance of being that value giving a zero shift with temperature change. The nearest 5% standard value 1200 ohms proved adequate for the production versions of the circuit.

7. Conclusion

A NaI photomultiplier gamma detector, fitted with an alpha source supplemented with a thermistor, and with appropriate electronics can be stabilized against changes in temperature, as well as the other causes of NaI photomultiplier gain drift.

The NaI p.m. detectors outfitted with both alpha source and thermistor are not significantly inferior to ordinary NaI p.m. detectors in resolution. Though they are 2 to 3 times more expensive than the equivalent size of ordinary NaI p.m. detectors, this increase in cost is still a small fraction of the overall system cost.

The associated circuitry makes use of only standard components, require negligible additional power, and entails minimal additional cost.

The stabilization which results from the use of this method allows better measurements to be made under unfavorable ambient conditions, and even in laboratory conditions has advantages in that less time and effort need be spent on calibration and set-up.

Therefore, while originally designed for use in nuclear safeguards, it is thought that the stabilization scheme should be of wider interest, such as a environmental, nuclear medicine, chemical tracer studies, etc., in field or laboratory surroundings.

The servo described here has been adapted for use in several commercial instruments, perhaps most notably mineature multichannel analyzers, intended for field use.

8. Acknowledgements

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I am indebted to Mr. Don Davidson (D.S. Davidson Co., North Haven, Connecticut) for information on the performance of the servo as he adopted it to the MCA developed by his firm.

References

1. The use of ^{241}Am alpha sources for this purpose was pioneered by the Harshaw Chemical Company, Solon Ohio; U.S. Patent #3030509, 1962.

2. Perhaps the earliest commercially successful application was the Eberline SAM I (for stabilized assay meter) widely used for gamma assay under field or factory conditions; improved versions (SAM II, etc.) are still available from Eberline, Santa Fe, New Mexico.

3. See for example: F. T. Kuchnir and F. J. Lynch IEEE Trans. Nucl. Sci. NS-15(3): 107 (1968).

M. L. Rousch, M. A. Wilson, and W. F. Hornyak Nucl. Instr. and Meth. 31(1): 112 (1964).

G. F. Knoll "Radiation Detection and Measurement" John Wiley & Sons, p.253.

4. C. V. Crannell, R. J. Kurz, and W. Viehmann Nucl. Instr. and Meth. 115, 253 (1974).

5. The so-called Brookhaven Survey Assay Meter, BSAM, circa 1977-79. A commercial version is produced by the IRT Corporation, San Diego, CA.

6. The light emitting diode light output is itself sensitive (amplitude and color) to temperature change; in any case, as regards the crystal, it can only warn about changes in opacity.

Illustrations