Plastic gamma sensors: an application in detection of radioisotopes

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ABSTRACT

A brief survey of plastic scintillators for various radiation measurement applications is presented here. The utility of plastic scintillators for practical applications such as gamma radiation monitoring, real-time radioisotope detection and screening is evaluated in laboratory and field measurements. This study also reports results of Monte Carlo-type predictive responses of common plastic scintillators in gamma and neutron radiation fields. Small-size plastic detectors are evaluated for static and dynamic gamma-ray detection sensitivity of selected radiation sources.

Keyword list
Scintillators, plastic, MCNP, gamma radiation, detection

1. INTRODUCTION

Organic scintillators in the form of solid, machinable plastic materials have long been an inexpensive source of radiation detector material. Plastic scintillators were first produced around 1950 in order to extend the developing liquid scintillator technology to that of a solid-phase medium. Plastic scintillators are characterized by the presence of a benzene ring structure in the constituent molecules. Plastic scintillators are solid solutions consisting of organic fluorescent compounds dissolved in a solidified polymer matrix. Most of the plastics used for light scintillation purposes are polystyrene (polyvinylbenzene) and polyvinyltoluene (polymethylstyrene). A variety of plastic scintillators has been developed to meet detection requirements[1]. Two plastic scintillators manufactured by Saint-Gobain Corporation, BC-400 and BC-404, have been in extensive general-purpose use for more than three decades. The BC-408, BC-412 and BC-416 plastic scintillators have almost 100% transparency for their own scintillation light and are used when the length of the scintillator exceeds 75 centimeters (cm) (30 inches [in]). The BC-454 contains a high concentration of enriched $^{10}B_5$ for thermal neutron detection purposes. Light output of 5% boron-loaded BC-454 is 48% that of anthracene. A slow neutron reacts with the boron inside the plastic material in the following way: $^{10}B_5 + n \rightarrow {}^{7}Li_3 + {}^{4}He_2$. The BC-418, BC-420 and BC-422 plastics have exceptionally short scintillation time constants, making them particularly useful for counting at extremely high rates or for high-precision coincidence counting. The BC-428 and BC-430 plastic scintillators have fluorescent spectra at longer wavelengths than do the other scintillators for spectral light collection applications. The BC-434 and BC-438 plastic scintillators have been formulated for use at higher temperatures. Some of the plastic scintillators contain lead or tin to enhance sensitivity to x-rays while another special scintillator, BC-470, is formulated to have a flat response to x-rays and gamma rays, making it useful for dosimetry. Two new developments are BC-444, which is unique among the many plastic scintillators in that its fluorescence decay time is relatively long, and BC-454, which contains boron for special neutron detection purposes. Most of these plastic scintillators have narrow areas of application; the general-purpose types such as BC-400, BC-404 and BC-408 comprise more than 80% of the plastic scintillators produced.

Large volumes of radiation-hardened plastic scintillators are used as charged-particle calorimeters for hadron physics. The Thomas Jefferson Laboratory has chosen 4-meter (m) (13.1-foot [ft])-long BC-412 plastic plates to build the calorimeter for the Large Acceptance Spectrometer[2]. The fast rise time (2-3 nanoseconds [ns]) of the scintillating pulses makes plastic scintillators attractive for timing purposes. Timing resolutions of a few hundred picoseconds are routinely achievable. Thick (20-cm [7.9-in]) "walls" of large-area (1 m^2 [10.74 ft^2]) plastic scintillators are used to detect energetic neutrons via knockout reactions; namely, $^{1}H(n, p)$ and 12C(n, p) taking place inside the volume of the plastic scintillator[3]. Recently, compact

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neutron coincidence spectrometers have been patented.[4] The coincidence unit is made up of a 1000 cm^3 (60.9 in^3) of plastic scintillator cylinder segmented and separated equally by three ^{6}Li-glass plates. Signals from the ^{6}Li plate represent thermal neutrons and indicate absorption of the energetic neutron in one of the four plastic segments. Because of low density and low atomic number, plastics cannot be used for energy spectrometry, and their intrinsic efficiencies are somewhat lower than those of inorganic scintillators. However, taking advantage of the large sizes and variety of shapes available, plastic scintillators are used for many gross gamma counting applications. They are extensively used as active sensor elements in walk-through and vehicle portal monitors for contamination detection and nuclear safeguard applications. Other uses include factory waste survey systems, laundry monitors and whole-body counters. Gamma portal monitors, typically used at a nuclear facility exit gate where all people either enter or leave the facility, use plastic scintillators to identify any inadvertent passage of radionuclides emanating gamma rays.

1.1 Fast neutron detector

The BC-720 is a scintillator designed specifically for detecting fast neutrons while rejecting gamma radiation in a mixed radiation field. It consists of zinc sulfide (silver) (ZnS[Ag]) phosphor embedded in a clear hydrogenous plastic material; it functions by means of a proton recoil interaction in the scintillator, the proton being detected by the zinc sulfide. The detector is a 15.9-millimeter- (mm) (0.62-in)-thick plastic disc, which can be mounted directly to photomultiplier tubes or to light guides using a variety of optical greases or epoxies. It is best used with photomultiplier tubes with high blue sensitivities such as those having bi-alkali, S-20 or S-11 type photocathodes. The gamma discrimination capability provided by the BC-720 is of particular value. The gamma pulse height is usually less than the neutron pulse height; in gamma fields below 1R/hr, the gamma rays can be easily rejected by pulse height discrimination. However, due to the random generation of recoil protons throughout the detector, the neutron spectrum is quite broad. In high gamma fields, the simultaneous detection of two or more gamma rays could produce pulse heights falling within the lower energy portion of the neutron spectrum; the use of a lower discriminator for gamma rejection would reduce the neutron detection efficiency. Gamma rejection may also be achieved by the use of time constants of a few microseconds.

1.2 Plastic scintillator as a new tool for spin physics

Polarized scintillating targets are now routinely available[5]. New possibilities for the measurement of spin-dependent observables in nuclear and particle physics are offered by the development of polarizable plastic scintillators. A polarized scintillating target is an instrument in which the hydrogen nuclei in a piece of scintillator can be dynamically polarized at very low temperatures and the light produced in the scintillator by scattered particles can be forwarded to a photomultiplier at room temperature.

1.3 Plastic shielding rings

Ultra-high-sensitivity detector systems are achieved by employing plastic scintillators as anticoincidence shields to reduce cosmic and terrestrial background. The shields often take the shape of annuli, wells or boxes. Similarly, the shields are used to increase the energy resolution of the detectors placed inside them.

1.4 Alpha and beta detection

Thin sheets and films of BC-400 and BC-434 are routinely used in survey instruments and for reactor gas monitors. Thin films are also used in beta dose detectors since the plastic materials are tissue equivalent to beta particles. Thus films and tiny spheres are used in constructing flow cells for monitoring beta activity in reactor cooling water. Since alpha particles have a limited range in air (approximately 2.5 cm [1 in]), a typical application, such as the Flow-Through Alpha Monitor[b] would use a detector with multiple (~16) plates. Using multiple plates has two other important advantages. First, it provides built-in redundancy. If one plate were to fail, only a portion of the detector would cease operation. Second, multiple plates allow one to distinguish between alpha events and natural background such as cosmic rays. The scintillating plates are made of a common plastic base, with a special scintillating chemical additive. For alpha detection, the Flow-Through Alpha Monitor system uses sheets of scintillator that are 30.5 x 30.5 cm (12 x 12 in) with a thickness of 1 mm (0.04 in) or a detector area of 1860 cm^2 (2 ft^2) on a single plate. One can add to the detector plates that are optimized for detecting more penetrating forms of radiation, such as beta or gamma rays. These plates are thicker, typically 13 mm (0.51 in).

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[b] Flow-Through Alpha Monitor, developed by Los Alamos National Laboratory in 1996, is a plastic detector system for monitoring alpha particles in a gas flow.

1.5 Radiation resistance of plastic scintillators

To examine the radiation hardness of plastic materials used in large hadron calorimeters, the Zeus experiment group irradiated small plastic samples with gamma rays and neutrons at external locations (HMI Berlin, GKSS-Research Reactor Geesthacht).[6] The radiation damage in polymeric material, often found to be receding, is somehow linked to the diffusion of oxygen. The Zeus group at CERN (European Organization for Nuclear Research) developed an apparatus especially to examine these effects. The group has developed a diffusion model, which described the recession of radiation damage during and after irradiation and was able to make quantitative predictions of radiation damage in the Zeus detector during ten years of operation. The working group was able to prove that irradiation produced short-lived absorption centers in some scintillator materials, which disappeared after a few hours. These absorption centers did not cause permanent radiation damage; however, recalibration of the detector was necessary.

2. PROPERTIES OF SCINTILLATORS

Inorganic crystalline scintillators such as sodium iodide (NaI), bismuth germanate, $Bi_4Ge_3O_{12}$ (BGO) and cesium iodide (CsI) are conventionally used in gamma-ray spectroscopy for their linear response, enormous light output (a factor of 4 better than a plastic scintillator), relatively higher absolute scintillation efficiency (~12%) and good energy resolution. Plastic scintillators have so far been ignored in field applications for gamma-ray measurements, even though they have some practical advantages over inorganic scintillators in terms of being less dense, less expensive, less temperature sensitive, rugged and are manufactured in large machinable volume. Plastic scintillators enjoy the unique advantage over NaI(Tl) that they have usable neutron response. The neutron detection efficiency (~8-12%) is dependent on the energy, threshold, thickness and volume of the plastic scintillator used. Neutron pulses are slower than gamma scintillator pulses and are easily separable in liquid scintillators. Characteristic physical properties of some common scintillator materials are listed in Table 1 below.

Table 1: Properties of scintillators

Material	Density (g/cc)	Wavelength of Maximum Emission	Refractive Index	Principal Decay Constant (m-Sec)	Total Light Yield	Pulse Rise Time (m-Sec)	Absolute Scintillation Efficiency	Relative Pulse Height
NaI(Tl)	3.67	415	1.85	0.23	38000	0.5	11.3	1.00
CsI(Tl)	4.51	540	1.80	1.0	52000	4.0	11.9	0.49
LiI(Eu)	4.08	470	1.96	1.4	11000		2.8	0.23
BGO	7.13	505	2.15	0.30	8200	0.8	2.1	0.13
BC-400	1.032	423	1.58	0.002	10000		3.0	0.25
BC-454	1.026	425	1.58	0.0022	10000		3.0	0.25

The ideal scintillator should possess the following properties:

- It should convert the kinetic energy of charged particles into detectable light with high scintillation efficiency.
- The conversion should be linear; the light yield should be proportional to deposited energy over as wide a range as possible.
- The medium should be transparent to the wavelength of its own emission for good light collection.
- The decay time of the induced luminescence should be short so that fast signal pulses can be generated.
- The material should be of good optical quality and subject to manufacture in sizes large enough to be of interest as a practical detector.
- Its index of refraction should be near that of glass (~ 1.5) to permit efficient coupling of the scintillation light to a photomultiplier tube.

Light output

Only a small fraction of the kinetic energy lost by a charged particle in a scintillator is converted into fluorescent light. The remainder is dissipated nonradiatively, primarily in the form of lattice vibrations and heat. The fraction of energy that is converted into fluorescence energy (scintillation efficiency) depends on the particle type and its energy. For organic scintillators such as anthracene, stilbene and other commercially available plastic scintillators, the response to electrons is linear for energies above 125 keV. The response to heavy charged particle like protons or alpha particles is always less for equivalent energies and is nonlinear to much higher initial energies. The light output dependence on the nature of the charged particle and the energy is shown in Figure 1. The light output of a scintillator depends on its efficiency for conversion of ionization energy to photons. The light output determines the efficiency and resolution of the scintillator. In general light output is different for different types of particles at the same energy. A given particle type does not always vary linearly with energy. Average energy losses required for creation of a photon for different materials by electron excitation are listed in Table 2.

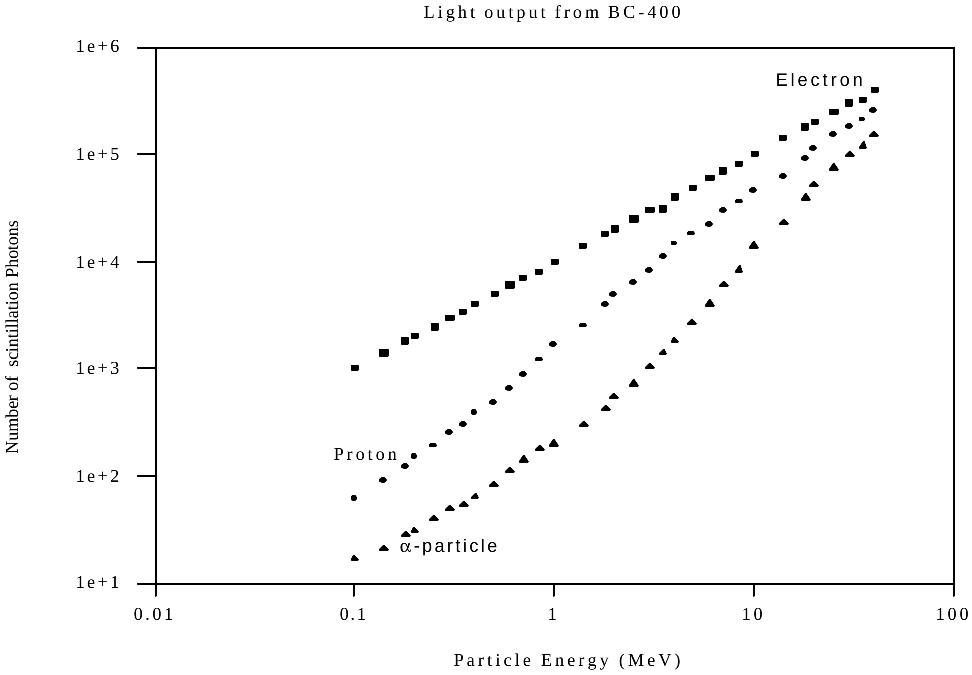

Figure 1. Light output of electrons, protons and α-particles in plastic scintillator as a function of incident particle energy

Table 2: Average energy loss per scintillator photon for electrons	
Materials	**eV/Photons**
Anthracene	60
NaI	25
BC-400	100
BGO	300

2.2 Efficiency

Two types of efficiencies relate to the discussion of gamma-ray detectors. Absolute efficiency is the ratio of the number of pulses recorded to the number of radiation quanta emitted by source. Intrinsic efficiency is defined as the ratio of the number of pulses recorded to the number of quanta incident on the detector. For an isotropic source, the two efficiencies are related by a solid angle subtended by the detector at the source.

$$\varepsilon_{int} = (4\,\pi/\Omega)\varepsilon_{abs}$$

where

Ω is the solid angle of the detector at the source. The intrinsic total gamma detection efficiency of a piece of BC-400 plastic scintillator of dimensions 50.8 x 76.3 mm (2 x 3 in) is shown in Figure 2 for different gamma-ray energies. The efficiency plots are shown for various distances between the source and the detector. The gamma rays are incident perpendicular to the detector surface.

To describe a relative percentile efficiency of gamma detection of plastic scintillators compared to that of NaI(Tl), one defines a term $A_{eff} = 100.(1 - \exp.(-\mu.\ L))$, where A is relative efficiency, μ is linear attenuation constant for the specific materials and L is the length traveled inside the specific medium. See Table 3.

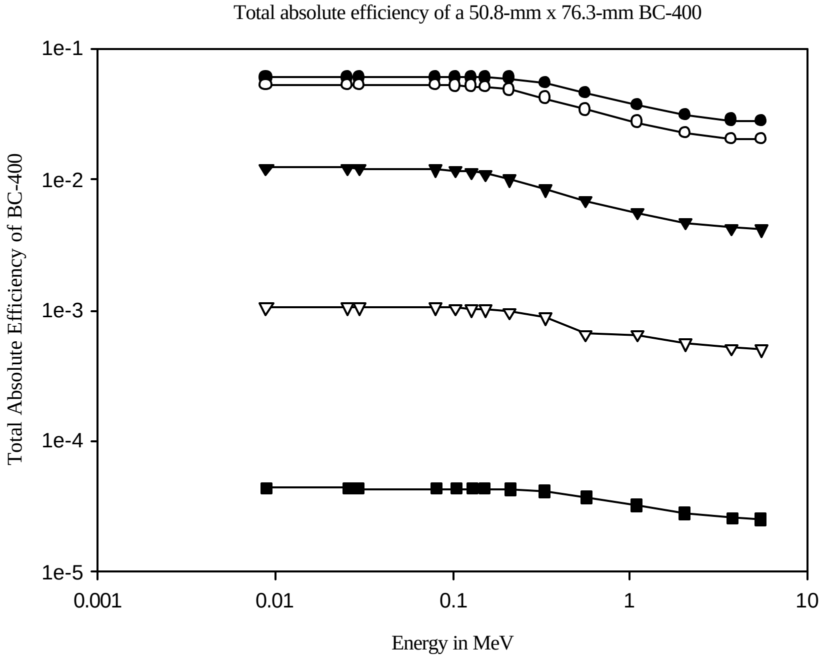

Figure 2. MCNP predicted total absolute gamma counting efficiency of a 50.8- x 76.2-mm (2- x 3-in) cylindrical BC-400 plastic scintillator. From top to bottom, the curves represent the efficiency at head-on distances of 0.0, 5.0, 10.0, 20.0 and 100.0 cm (0.0, 1.96, 3.93, 7.86 and 39.3 in) from the source.

Table 3: Required distance traveled (L, cm) as a function of γ-energy (E_{γ}, MeV) for selected relative efficiency (A_{eff})

A_{eff}	50 %		70 %		90 %	
E_{γ}	L_{NaI}	L_{pl}	L_{NaI}	L_{pl}	L_{NaI}	L_{pl}
0.08	0.07	3.8	0.11	6.7	0.21	12.7
0.10	0.12	4.0	0.21	7.0	0.39	13.4
0.20	0.61	4.9	1.1	8.5	2.0	16.2
0.30	1.2	5.6	2.1	9.7	4.0	18.6
0.40	1.7	6.3	2.9	10.9	5.6	20.8
0.50	2.1	6.8	3.6	11.9	6.8	22.7
0.60	2.4	7.4	4.1	12.9	7.8	24.6
0.80	2.9	8.4	5.0	14.7	9.5	28.0
1.0	3.3	9.4	5.7	16.3	10.8	31.2
1.5	4.1	12	7.0	20.1	13.5	38.4
2.0	4.6	14	8.0	23.5	15.2	45.0
3.0	5.2	17	9.0	29.9	17.2	57.1

It is noteworthy that for the same values of A_{eff}, the ratio, $L_{NaI}/L_{pl,}$ increases with increasing values of E_γ and is 0.27 at 400 keV. In the energy region of interest (60 through 3000 keV), the gamma-ray detection efficiency of plastic scintillators relative to that of NaI can be approximately given by $(30 \pm 3)\%$.

The gamma detection efficiency of organic phosphors in the energy region between 50 keV and 3 MeV is dominated by a Compton interaction. When applied to gamma-ray detectors, one defines absolute efficiency as the ratio of the number of pulses recorded to the number of radiation quanta emitted by the source. The intrinsic efficiency is defined as the ratio of the number of pulses recorded to the number of quanta incident on the detector. For an isotropic source, the two efficiencies are related by a solid angle subtended by the detector at the source. The absolute total efficiencies for a 50.8- x 76.2-mm (2 x 3 in) BC-400 cylinder at different gamma-ray energies are shown in Figure 2 for various distances between the source and the detector.

3. MCNP WORK

The Monte Carlo N-Particle (MCNP[7]) is a general-purpose neutron, photon and electron transport code developed at Los Alamos National Laboratory. MCNP incorporates all three major photo interactions, namely, photoelectric effect, pair production and the most dominant effect in plastic scintillator, the Compton effect. The mean free path is calculated as

$$\lambda = 1/\rho\sigma$$

Where ρ is the atomic density and σ is the reaction cross section and can be decomposed as

$$\sigma = \sigma_{Comp} + \sigma_{Photo\text{-}electric} + \sigma_{pair\text{-}production}$$

The calculated linear gamma-ray attenuation coefficient of BC-400 is compared with the nominal values quoted by Saint-Gobain Corporation in Figure 3. The gamma-ray pulse height spectrum from BC-400, as calculated by MCNP, is compared to experimental measurements in Figure 4.

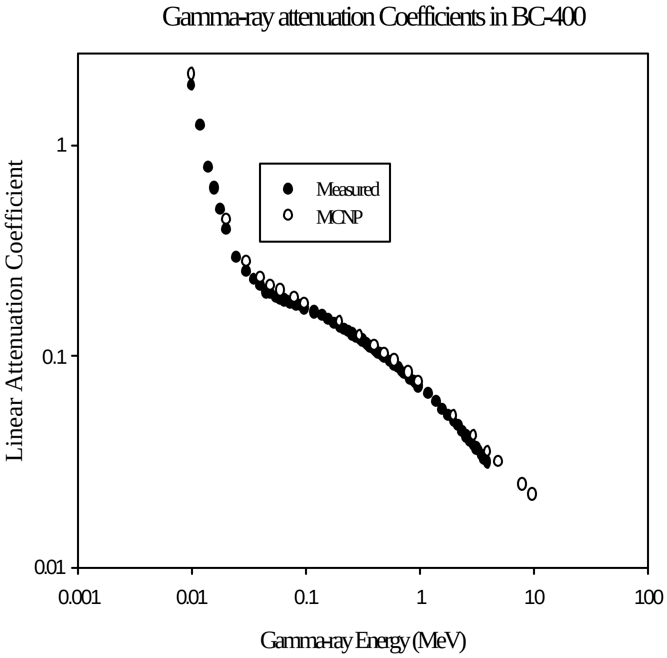

Figure 3. Linear gamma-ray attenuation coefficients for the BC-400 plastic scintillator are computed using MCNP and compared to the data published by Bicron Corporation.

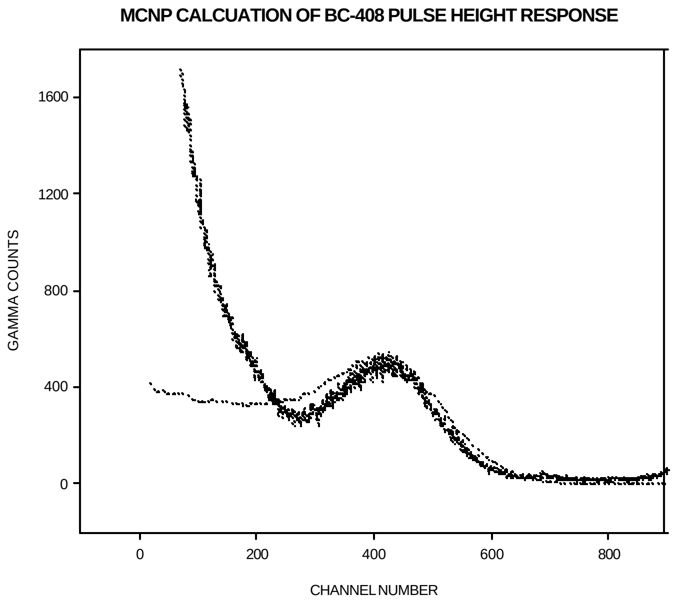

Figure 4. MCNP calculations compared with experimental data.

Responses for the BC-454 plastic scintillators were determined by Monte Carlo calculations using MCNP code. The geometry of the scintillator was a 12.7-cm (5-in) cube. The scintillator was modeled for ^{10}B neutron absorption and proton recoil events. The neutron absorption runs were conducted over the range of energy of a thermal neutron to 1 MeV mono-energetic neutron sources. Absorptions were based on 5% substitution of natural boron (1% ^{10}B). The neutron energy deposited for several monoenergetic neutrons is shown in Figure 5.

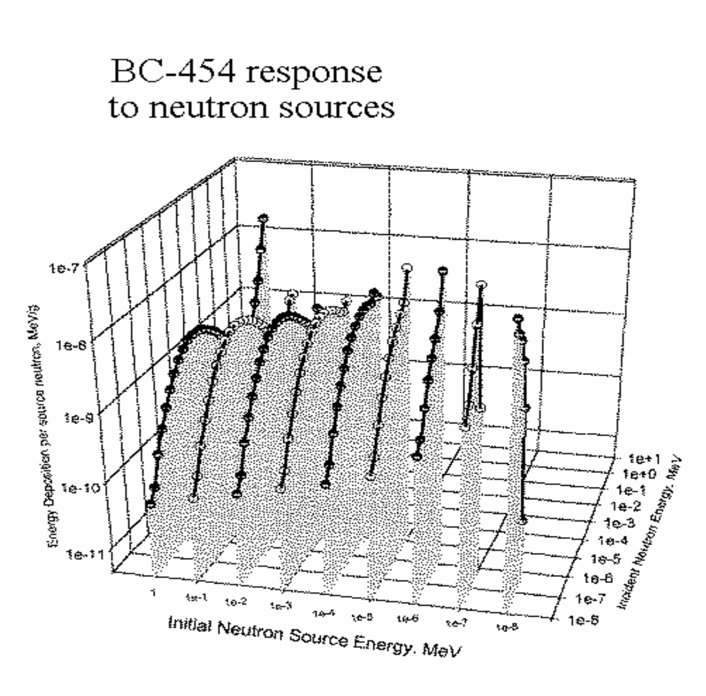

Figure 5. MCNP-predicted response of BC-454 for mono-energetic neutron sources. The energy deposited in the plastic scintillator is shown for various single-energy neutron beams. The source is placed at the center of a 3.05- x 3.05- x 3.05-m (10- x 10- x 10-ft) room with 0.305-m- (1-ft)-thick concrete walls. The response is for a 12.7- x 12.7- x 12.7-cm (5- x 5- x 5-in) BC-454 detector placed at the center of any wall.

4. EXPERIMENTAL WORK

Energy calibration of a plastic gamma sensor can be made using the Compton edges of several mono-energetic gamma sources. The Compton edges as shown in Figure 6 can be defined as

$$T_{CE} = E_\gamma^2 / (255.5 + E_\gamma) \text{ in keV}$$

where E_γ is the incident gamma energy.

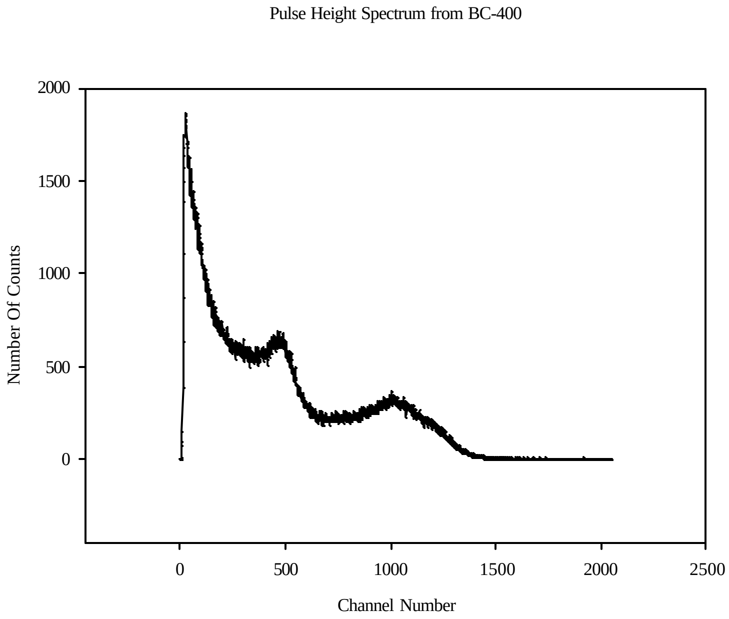

Figure 6. A typical gamma-ray pulse height spectrum from BC-400. Due to the poor energy resolution in plastic scintillators, the Compton edges for the two closely spaced γ-lines from ^{60}Co-isotope overlap.

The photo-response of a plastic scintillator to incident electrons is linear with electron energy above 125 keV and can be expressed in the linear region as P (E) = c (T_{CE} - E_0), where P (E) is the relative pulse height, T_{CE} is the maximum recoil Compton electron energy and E is the incident gamma energy. E_0 is the energy intercept and c a scaling parameter. A plot of T_{CE} for different gamma sources versus pulse height channel numbers is a straight line that can be used to calibrate the scintillator light output as shown in Figure 7. T_{CE}, the parameter used in detector calibration, can be ascertained accurately from the measured coincidence gamma spectrum.

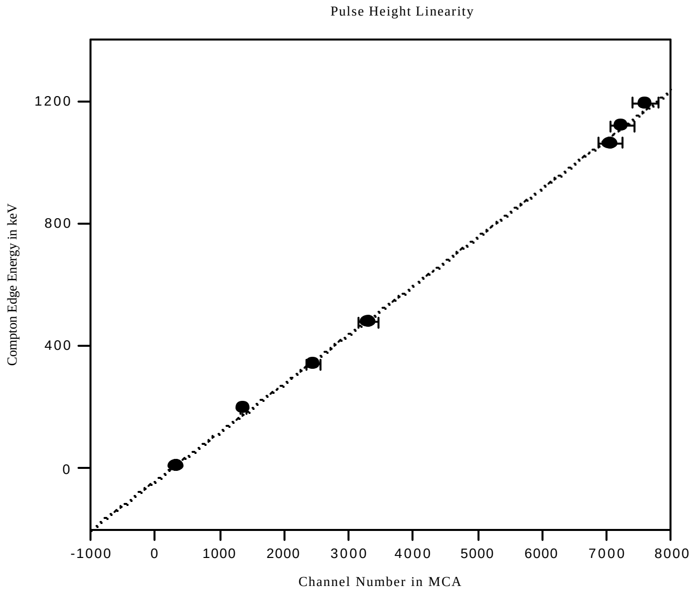

Figure 7. Multichannel analyzer calibration plot for BC-400 shows some non-linearity in the high-energy range.

The plastic scintillator BC-400 was in the form of a cylinder. The angular response of the detector was measured carefully by counting the gamma rays from spectral analysis at different angles (with respect to the detector axis of symmetry). The response shows cylindrical symmetry and it is forward peaked, as shown in Figure 8.

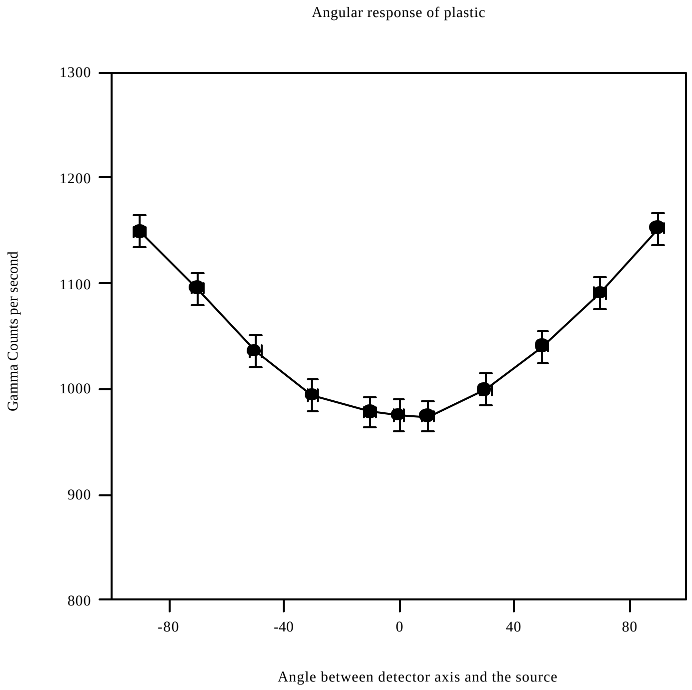

Figure 8. The angular distribution of gamma counts is seen by the BC-400 plastic scintillator from a button source. The angles are measured with respect to the cylindrical axis of the detector.

Experimental studies have been conducted to compare the relative efficiency of plastic scintillators compared to that of NaI(Tl) and CsI(Tl) crystals of the same size in static measurements. Gamma-ray count rates in the presence of a button source of radionuclide ^{137}Cs (< 18.5 KBq) at a distance of 25.4 cm (10 in) along the cylindrical axes of three detectors are shown in Figure 9.

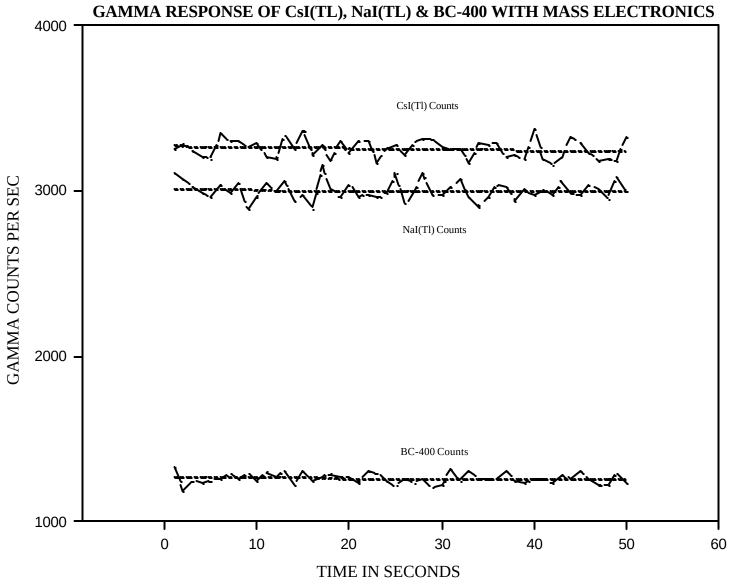

Figure 9. Gross count rates from 50.8- x 76.2-mm (2- x 3-in) inorganic crystals are compared to those from a BC-400 plastic scintillator.

The mobile data acquisition electronics, developed by EG&G, Inc. and used in the above measurements, are ideally suited to locate point radiation sources. The sensor system can be configured according to the detection platforms. Using the standard algorithm for normal point source dynamic detection procedures, the final calculated outputs are compared for a 50.8- x 76.2-mm (2- x 3-in) cylindrical plastic scintillator BC-408 and NaI(Tl) crystal. The algorithm output plotted in Figure 10 compares the measured gamma source counts with respect to the variation of the background as a ^{137}Cs source is moved from a distance. The peak for each detector response corresponds to the point of closest approach of the source to the detector.

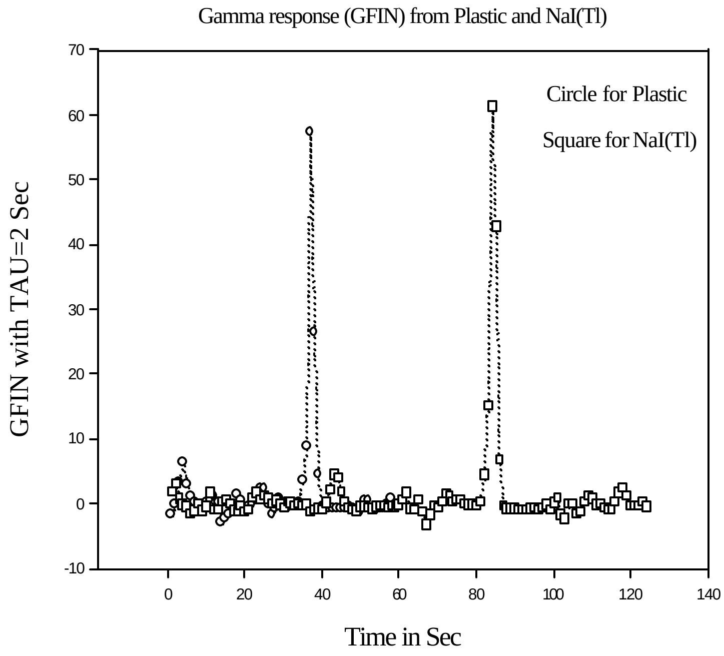

Figure 10. Comparison of algorithm output from a 50.8- x 76.2-mm (2- x 3-in) cylindrical crystal of NaI(Tl) and a plastic BC-408 scintillator.

5. CONCLUSION

Plastic scintillators are used in multiple radiation detection and measurement systems under diverse experimental conditions, because of their light weight, large area, considerable light output, radiation hardness and polarizability. For safeguard applications, plastic scintillators can be used in portal and vehicle monitors. Even though plastic scintillators have significant non-linearity in their light output (so no spectral information can be obtained from gamma radiation) for non-spectroscopic gamma detection work, plastic scintillators are better than inorganic scintillators.

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