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A LaC₁₃(Ce) Scintillator, And An NaI(Tl)
Scintillator**

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Performance comparison of four compact room-temperature detectors – two cadmium zinc telluride (CZT) semiconductor detectors, a $\text{LaCl}_3(\text{Ce})$ scintillator, and an $\text{NaI}(\text{Tl})$ scintillator

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Abstract— The performance characteristics of four compact, room-temperature detectors – two scintillators and two semiconductor detectors – were studied. All are commercially-available detectors. The two scintillators were a $\text{Ø}13$ mm X 13mm lanthanum chloride [$\text{LaCl}_3(\text{Ce})$] detector and a $\text{Ø}25$ mm X 25 mm sodium iodide [$\text{NaI}(\text{Tl})$] detector. The two semiconductor detectors were a 10 mm X 10 mm X 3 mm cadmium zinc telluride (CZT) detector with a coplanar gridded anode and a 5 mm X 5 mm X 5 mm CZT detector with an extended cathode. The efficiency, resolution, and peak shape of these devices are compared. Since $\text{LaCl}_3(\text{Ce})$ is a relatively new commercial scintillator material, additional information on the performance of this detector is presented. Specifically, the impact of naturally-occurring radioactive ^{138}La and additional contamination from alpha-emitting radionuclides on the background measured with this detector are discussed.

Index Terms— Gamma-ray spectroscopy detectors, $\text{LaCl}_3(\text{Ce})$ scintillation detectors, Room temperature semiconductor detectors

I. INTRODUCTION

While High Purity Germanium (HPGe) detectors, with their unexcelled gamma-ray energy resolution, are often the best choice for gamma-ray spectroscopy applications; the requirement for cryogenic cooling during operation complicates their use in certain applications. This work compares the detection efficiency and resolution capabilities of four alternative gamma-ray spectrometric detectors that operate at room temperature. Although none of these units provides the combination of resolution and detection efficiency afforded by a HPGe, they have performance

features that make them attractive for certain radiation measurement scenarios where portability, compact design, and freedom from cryogenics are primary considerations.

A recent publication provided performance comparison data on three detectors including a $\text{Ø}10$ cm X 10 cm $\text{NaI}(\text{Tl})$, a $\text{Ø}55$ mm X 54 mm HPGe detector, and a $\text{Ø}9$ mm X 2 mm CdTe detector [1]. Our work, reported in this paper, provides complimentary assessment data on a different suite of four highly portable detectors, including work with the promising new scintillator $\text{LaCl}_3(\text{Ce})$.

II. EXPERIMENTAL METHOD

The electronics system used for these evaluations consisted of a preamplifier, a linear amplifier (Tennelec TC244 or Ortec 460), and an multichannel analyzer (Amptek MCA8000A) along with the required high voltage bias supply (ORTEC 556 or ORTEC 459). Amplifier pulse shaping times and system gains were adjusted to match the requirements of the detector and the desired peak positioning. All spectra were stored in 2048 channels.

A set of three sealed point sources were used for this evaluation. The source radionuclides, their activities and estimated total uncertainties on 01-January-04 were ^{57}Co (1.95 ± 0.09 kBq), ^{60}Co (117 ± 5 kBq), and ^{137}Cs (360 ± 11 kBq). Source-to-detector distances were 10.0 ± 0.2 cm unless otherwise noted. The system gain for each detector was adjusted to position like peaks in the same channel. Thus the spectra of ^{57}Co taken with all four detectors had the 122 keV peak in channel 368 ± 3 , the ^{137}Cs 662 keV peak in channel 976 ± 5 and the ^{60}Co 1332 keV peak in channel 1737 ± 3 .

Two cadmium zinc telluride (CZT) detectors were evaluated in this study. The first (designated detector C5) was a 5 mm X 5 mm X 5 mm (125 cm^3) crystal obtained from eV Products. This detector is a quasi-hemispherical device with a full area anode and extended cathode that provides improved charge collection characteristics and thus improved resolution and peak shape relative to a pure planar design [2]. The

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detector is packaged along with a hybrid preamplifier in an integral package $\varnothing 12$ mm by 89 mm. During testing the detector bias voltage was +1000 V. The amplifier shaping time was 0.375 μ sec. Testing determined that the face of the detector was 13 ± 2 mm back from the detector end cap. Thus, to achieve a source-to-detector distance of 10.0 cm, the ^{57}Co and ^{137}Cs sources were positioned 8.7 cm from the detector C5 end cap. The ^{60}Co source was counted at a 5 cm source-to-detector distance or 3.7 cm from the end cap; and the data corrected to a source-to-detector distance of 10 cm using an inverse square assumption.

The second CZT detector (designated CP) was a 10 mm X 10 mm X 3 mm (300 mm^3) detector obtained from Spire Corporation. This crystal was equipped with a coplanar-gridded anode to significantly improve the unit's resolution performance [3]. The detector was packaged along with the required preamplifiers and analog subtraction circuit in a $\varnothing 3.2$ cm by 13 cm unit. The detector was operated with a bias voltage of -600 V and a grid bias of -60 V. The amplifier shaping time was 0.375 μ sec. All sources were counted at a distance of 9 cm from the detector end cap. However, testing indicated that the detector face was located 1.8 ± 0.4 cm back from the anodized aluminum end cap. Consequently, sources counted at a source-to-end cap distance of 9 cm were actually 10.8 ± 0.4 cm from the detector. The results for this detector presented in this report have been adjusted to a source-to-detector distance of 10 cm by the inverse square law.

Two scintillation detectors were included in this evaluation – a $\text{LaCl}_3(\text{Ce})$ and a $\text{NaI}(\text{Tl})$ detector. The $\text{LaCl}_3(\text{Ce})$ detector (designated L2) was a $\varnothing 13$ mm X 13 mm (1.7 cm^3) crystal of LaCl_3 with 10% Ce. This scintillation material is reported to have an excellent combination of scintillation light output and resolution [4]. The detector was an integral unit coupled to a Photonis XP1911 photomultiplier tube with a dividing network configured for positive high voltage. A high voltage setting of +800 V was used. Some spectra were acquired using a delay line amplifier (ORTEC 460) and a 0.25 μ sec shaping time. Other spectra used the TC244 amplifier and a shaping time of either 0.375 or 6 μ sec. All sources were counted at a source-to-detector distance of 10 cm.

In addition to the source spectra acquired, a number of background spectra were taken with the $\text{LaCl}_3(\text{Ce})$ detector. Background spectra were of particular interest with this detector for two reasons. First, Lanthanum has a naturally-occurring radioactive isotope, ^{138}La (1.05×10^{11} year half-life) present in an abundance of about 0.09% in natural La and we wanted to assess the effect of this ^{138}La on the crystal background. Second, during preliminary work with this scintillator, we and another researcher [5] noted a puzzling set of broad peaks in the background at energies between about 1500 and 3000 keV. We suspected that these peaks were from alpha particle emitter contamination in the $\text{LaCl}_3(\text{Ce})$ crystal and wanted to investigate.

The second scintillation detector (designated N1) was a fairly standard $\varnothing 2.54$ cm X 2.54 cm $\text{NaI}(\text{Tl})$ detector manufactured by Teledyne Isotopes, Inc. The detector was an integral unit mated to a 5.08 cm diameter photomultiplier. The base was an Ortec Model 266 and the AC-coupled dynode output signal was used directly as input to the TC244 amplifier. The high voltage was set to +780 V. The amplifier shaping time was 1.5 μ sec.

The spectra were acquired using the Amptek program PMCA Version 2.01 specifically provided for control of the 8000A Pocket MCA. Most net photopeak areas were computed using a simple region-of-interest summation with a stepped background subtraction, and photopeak resolution values (FWHM) were computed using analytical interpolation of the channel contents, an approach that provides a better and more consistent measure of the overall peak widths when photopeak shapes are non-Gaussian than do FWHM values derived from peak fitting results. An exception to this approach was for the area calculation of the 122 keV peak of ^{57}Co in the spectra acquired with the two scintillation detectors. The resolution of the L2 and N1 detectors was insufficient to resolve the 122 keV and the 136 keV peaks. Consequently, the area of the 122 keV peak in these spectra was computed by non-linear least squares fitting of the 122-136 keV peak region as a doublet with Gaussian functions and a step background.

III. RESULTS AND DISCUSSION

Table 1 compares the measured efficiency in counts per emitted gamma-ray at a 10 cm source-to-detector distance, and the resolution at half maximum height (FWHM) and the full width at one-tenth maximum height (FWTM) of the four detectors for the major peaks of the three radionuclides counted. All uncertainties are quoted at one estimated standard deviation.

Additionally, Figure 1 plots the spectral region around the 662 keV peak for the ^{137}Cs point source at 10 cm from each detector. Not only does this figure illustrate the comparative efficiency and resolution of the detectors tested, it highlights the significant peak shape differences noted with certain of these detectors.

Only the $\text{NaI}(\text{Tl})$ detector N1 performed exactly as expected. Although the N1 resolution performance was the poorest of the four tested detectors, it exhibited the expected photopeak efficiency and peak shape. Details regarding the performance of the other three detectors are discussed below.

A. The 125 mm³ CZT detector (C5)

The least efficient (and most compact) of the detectors studied is the 125 mm³ eV Products detector (C5). This

PERFORMANCE PARAMETERS OF THE MEASURED PHOTOPEAK EFFICIENCY IN COUNTS PER EMITTED GAMMA-RAY AT A 10 CM SOURCE-TO-DETECTOR DISTANCE, AND THE RESOLUTION AT ONE- HALF (FWHM) AND AT ONE-TENTH MAXIMUM HEIGHT (FWTM) OF THE FOUR COMPACT DETECTORS FOR THE PEAKS OF INTEREST. ALL UNCERTAINTIES ARE QUOTED AT ONE ESTIMATED STANDARD DEVIATION.

Detector	Nuclide	Energy (keV)	Efficiency c/γ @10 cm	FWHM (keV)	FWHM (%)	FWTM (keV)	FWTM (%)	FWTM/FWHM Ratio
C5 CZT [125 mm ³]	⁵⁷ Co	122.1	(9.6±0.5)X10 ⁻⁰⁵	4.0 ± 0.1	(3.2±0.1)%	12 ± 1	(10 ± 1)%	3.1 ± 0.3
	¹³⁷ Cs	661.7	(1.92±0.06)X10 ⁻⁰⁶	16 ± 3	(2.4±0.4)%	77 ± 9	(12 ± 1)%	5 ± 1
	⁶⁰ Co	1332.5	(2.6±0.1)X10 ⁻⁰⁷	27 ± 2	(2.1±0.2)%	101 ± 8	(7.6 ± 0.6)%	3.7 ± 0.4
CP CPG CZT [300 mm ³]	⁵⁷ Co	122.1	(2.3±0.1)X10 ⁻⁰⁴	5.2 ± 0.3	(4.2±0.2)%	12 ± 1	(9.5 ± 0.9)%	2.2 ± 0.2
	¹³⁷ Cs	661.7	(7.0±0.2)X10 ⁻⁰⁶	13.7 ± 0.4	(2.07±0.06)%	57 ± 5	(8.7 ± 0.8)%	4.2 ± 0.4
	⁶⁰ Co	1332.5	(1.15±0.05)X10 ⁻⁰⁶	18 ± 1	(1.31±0.08)%	70 ± 4	(5.2 ± 0.3)%	4 ± 0.3
L2 LaCl ₃ (Ce) [Ø13 mm X 13 mm]	⁵⁷ Co	122.1	(6.2±0.3)X10 ⁻⁰⁴	11.9 ± 0.9	(9.8 ± 0.7)%	23 ± 4	(19 ± 3)%	1.9 ± 0.4
	¹³⁷ Cs	661.7	(4.7±0.1)X10 ⁻⁰⁵	27.5 ± 0.3	(4.15 ± 0.05)%	60 ± 5	(9.0 ± 0.8)%	2.2 ± 0.2
	⁶⁰ Co	1332.5	(1.34±0.06)X10 ⁻⁰⁵	48.5 ± 0.4	(3.64 ± 0.03)%	94 ± 7	(7.1 ± 0.5)%	1.9 ± 0.1
N1 NaI(Tl) [Ø25 mm X 25 mm]	⁵⁷ Co	122.1	(2.8±0.1)X10 ⁻⁰³	10.1 ± 0.2	(8.3±0.2)%	17 ± 2	(14 ± 2)%	1.7 ± 0.3
	¹³⁷ Cs	661.7	(4.3±0.1)X10 ⁻⁰⁴	42.7 ± 0.2	(6.45±0.03)%	78 ± 9	(12 ± 1)%	1.8 ± 0.2
	⁶⁰ Co	1332.5	(1.44±0.06)X10 ⁻⁰⁴	60.0 ± 0.5	(4.51±0.04)%	108 ± 9	(8.1 ± 0.7)%	1.8 ± 0.2

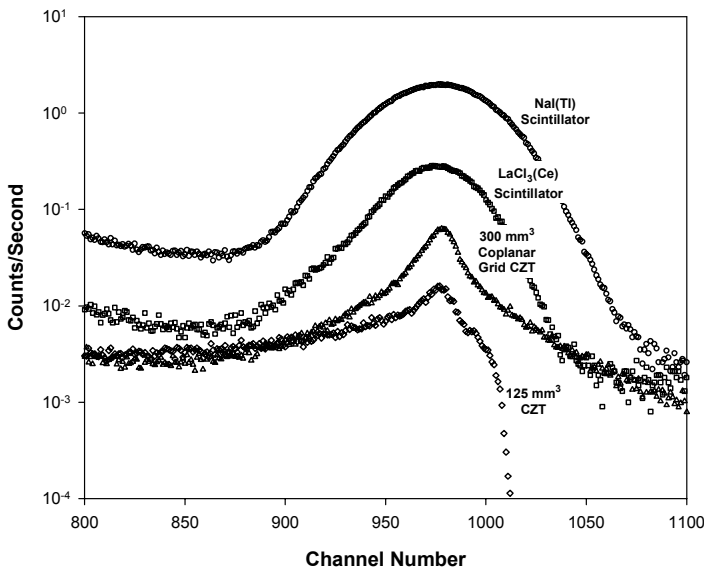


Fig. 1. Comparison of the 662 keV photopeaks acquired on the four tested detectors at a source-to-detector distance of 10 cm. This figure highlights the peak shape differences.

unit incorporates what the manufacturer calls CAPTURE^a technology that uses a full area anode and a cathode contact plated on the face and extended to some height up the sides of the CZT crystal [2]. This electrode pattern produces a quasi-electron only charge collection reducing the effect of poor hole mobility on the detector performance, and thus reducing the spectral peak tailing. Our laboratory has purchased, tested, and used six or more of these detectors, and has deployed them in spent fuel monitoring systems [6].

An odd feature of these detectors is the 662 keV peak shape apparent in Figure 1. While the long low energy tail is a common feature in spectra taken with room temperature semiconductor detectors, the pronounced high-energy shoulder

starting just below the half maximum height on the high-energy side of peaks acquired with these detectors is unusual.

The energy dependence of this shape perturbation was investigated. Spectra from ¹³³Ba and ¹⁵²Eu sources were acquired on three detectors of this type, and the peak shapes inspected. While there are some differences noted between detectors, spectral peak shapes for photopeaks with energies below about 200 keV, for instance the 81.0 keV line from ¹³³Ba and the 121.8 keV line from ¹⁵²Eu do not show this odd high-side shoulder, and are well fit by Gaussian functions. However, this perturbation is visible in most cases at 244 keV and in all cases at 300 keV and above.

In consultation with the manufacturer, the explanation for this odd peak shape is believed to be as follows. The electric field distribution of the CAPTURETM electrode geometry is non-uniform in nature. There is an optimum plating height for the extended cathode in these CAPTURETM detectors for a given photon energy, and the commercial devices are optimized for photon energies <150keV [2]. For irradiation through the cathode, photons of about 150 keV or less primarily interact within the detector near the cathode and under the cathode “CAP”. The electric field under the CAP cathode contact is of lower intensity than the field outside the CAP region, thus hole collection to the cathode is inhibited by the low field strength, and the device functions as a quasi electron-only collection device. However, photons with energies greater than 150 keV tend to interact throughout the small detector volume. Electric field maps of these CAPTURETM devices show small regions of very high field strength at the edge of the CAP electrode [2]. The high energy shoulder noted in spectra acquired with CAPTURETM detectors is probably due to these small high electric field strength regions near the CAP electrode edges. As a consequence of their high electric field strength and proximity to the cathode, these small regions will have a higher charge collection efficiency for both holes and electrons. The enhanced hole collection efficiency allows more total charge collection per event and will induce a slightly higher "effective gain" in this region than is present in the rest of the detector

^a This term is a trade mark of eV Products , a division of II-VI Inc. Saxonburg, PA, USA

crystal. The small number of photoelectric events that occur in this region would therefore be expected to show up slightly higher in the energy spectrum producing the high side shoulder seen in spectra of high-energy photons taken with CAPture™ detectors.

The strong low energy tail exhibited by this and similar detectors requires special calculational approaches in gamma-ray spectral fitting and analysis codes. Region-of-interest summation codes must either use stepped background functions [7] to compute the net area of the entire peak, or confine the area computation to the high energy half of each tailed peak [8]. Gamma-ray spectral fitting codes capable of joining tail functions, either as exponential or skewed Gaussian additions to the primary Gaussian functions, generally provide acceptable fits to CZT acquired peaks. For example, a version of our PCGAP code [9], HYPERMET-PC [10] and the commercial software package GENIE 2000 from Canberra industries can provide acceptable fits to the low energy tail and perform well on peaks without a high-side shoulder. However, neither of these analysis codes could properly fit the odd high side shapes exhibited by detector C5 above about 300 keV.

B. The 300 mm³ coplanar gridded CZT detector (CP)

The next most efficient detector is the 300 mm³ coplanar-gridded anode CZT detector. As a consequence of its larger size this detector is more efficient than the 125 mm³ CAPture™ detector (C5), and thanks to its gridded electrode arrangement has somewhat better FWHM resolution at 662 keV.

CZT detectors with coplanar-gridded anodes are popular room temperature detectors because they provide reasonable resolution performance along with moderate available volumes (on the order of 1 to 2 cm³). The principle of operation has been described in the literature [3].

The CPG CZT detector used in this work was a 10 mm X 10 mm X 3 mm device obtained from Spire Corporation in 1999. A prominent feature of spectra acquired with this detector is the unusual peak shape displayed in Figure 1. Although the FWHM at 662 keV is a respectable 2.07% ($\pm 0.06\%$), the FWTM/FWHM ratio for this peak is 4.2 ± 0.3 , a value more than twice that expected for a pure Gaussian. These base-broadened peaks are not well fit by standard analysis routines that implement only low side tailing. Analysis codes have been developed that fit these peaks to Gaussian functions with strong high- and low-side smoothed exponential tails [11] and the authors had good success fitting these peaks to a Lorentzian function using a standard graphical analysis package.

This highly base-broadened peak shape is not a characteristic of all CPG CZT detectors. Our CPG detector is about 6 years old. One reason for this broadening below half maximum has been reported to be the actual pattern used for the coplanar-gridded anode, and whether or not that pattern includes edge compensation [12]. Modern detectors of this type can have much more Gaussian peak shapes. For example,

at least one manufacturer is selling CPG CZT detectors with a volume of 2.25 cm³ (15 mm X 15 mm X 10 mm) with a resolution of 2.6% at 662 keV, and a FWTM/FWHM ratio of 1.97.

C. The LaCl₃(Ce) scintillation detector

Cerium-activated lanthanum halide scintillators are new scintillation materials that are reported to have very high light output, good resolution, excellent response linearity, and fast timing characteristics [4]. The detector tested in this work was a $\varnothing 13$ mm X 13 mm (1.7 cm³) crystal of LaCl₃(10% Ce). The measured resolution and efficiency values are presented in Table 1.

Natural lanthanum is 99.91% stable ¹³⁹La and 0.09% ¹³⁸La. Lanthanum-138 is naturally radioactive decaying through electron capture (EC) decay (66.7%) and beta decay (33.3%) with a half life of 1.06×10^{11} years [13]. Thus our $\varnothing 13$ mm X 13 mm LaCl₃(10% Ce) crystal would have a calculated 2.7 Bq of ¹³⁸La. The EC decay is to a 1435.8 keV excited state in ¹³⁸Ba, that de-excites through a single 1435.8 keV gamma transition. The less prevalent beta branch has a Q value of 1044 keV and leads to a 788.7 keV state in ¹³⁸Ce that then decays through a single gamma transition to the ground state. The consequences of the radiation from ¹³⁸La within the detector are expected to be (1) a Ba K X-ray peak at an energy of 32.9 keV (weighted average) from the EC decays in which the low energy X-rays interact and the 1435.8 keV gamma-ray escapes with no interaction in the crystal, (2) a peak at 1469 keV from events in which both the 1435.8 keV gamma ray and the 32.9 keV X-ray are fully stopped in the crystal, (3) a Compton continuum from the 1435 + X-ray interactions that scatter with part of their energy leaving the crystal, (4) a continuum from β^- decay followed by the 788.7 keV γ -ray transition that extends to the beta Q value of 1044 keV.

All of these features have been identified in background spectra taken with our L2 detector. However, unexpected broad peaks, suspected to be from α -particle emitting contaminants in the scintillation crystal, at electron-equivalent energies between about 1600 keV and 3000 keV dominate these background spectra. One of the authors had previous experience with a lanthanum compound that was intractably contaminated with low activity levels of 21.8-y ²²⁷Ac + daughters, thus we hypothesized this as the contaminant. An annotated spectrum is presented in Figure 2.

By counting the L2 detector crystal as a source on a low background gamma-ray spectrometer, and by synthesis of the expected alpha particle spectral shapes, we have identified the source of these peaks as contamination of the LaCl₃(Ce) crystal with ²²⁷Ac and its daughters. The activity of the ²²⁷Ac contaminant was determined by counting the L2 crystal on a low background gamma-ray spectrometer. The ²²⁷Ac content of the detector was determined from the net count rate of the 832 keV line of the ²¹¹Pb daughter. The detection efficiency was

estimated using the net count rate in the 788 keV gamma ray line of ^{138}La , and an emission rate for this gamma ray determined from the calculated content of natural ^{138}La in the crystal. Using this approach, we measured a ^{211}Pb activity (and thus an ^{227}Ac activity) of 1.3 ± 0.3 Bq.

Each decay of ^{227}Ac through its daughter chain generates 5 alpha particles. Thus 1.3 ± 0.3 Bq of ^{227}Ac decaying to its stable ^{207}Pb daughter would generate 6.5 ± 1.5 alpha emissions per second. The total count rate registered in the spurious peaks between 1500 and 3000 keV noted in the L2 $\text{LaCl}_3(\text{Ce})$ detector background is 5.7 ± 0.1 counts per second. This rate agrees very well with that expected from the measured amount of ^{227}Ac + daughters.

IV. CONCLUSIONS

This study has documented several performance parameters of a set of four compact room temperature detectors. The photopeak detection efficiency for point sources on axis at a source to detector distance of 10 cm and the resolution performance of a 125 mm³ CZT detector with a special CAPture™ electrode configuration, a 300 mm³ CZT detector with a co-planar gridded anode, a Ø13 mm X 13 mm $\text{LaCl}_3(\text{Ce})$ scintillation detector, and a Ø25 mm X 25 mm NaI(Tl) scintillation detector were measured and documented for gamma-ray energies of 122, 662, and 1332 keV. While none of these detectors have the resolution or detection efficiency performance of a germanium detector, their small size, relatively low cost, and moderate performance may dictate their use in some applications.

This work identified some additional characteristics of each of the tested detectors [other than the “tried and true” NaI(Tl) detector] that may limit their use in some applications. The two CZT detectors exhibited peak shapes at energies above about 200 keV that complicated analysis of spectra taken with these units. The 300 mm³ coplanar gridded CZT detector exhibited base-broadened peak shapes more Lorentzian than Gaussian in shape. Peaks from gamma rays with energies greater than about 250 keV acquired with the 125 mm³ CZT detector with CAPture™ electrodes exhibited a high-energy shoulder believed due to the charge collection characteristics of the CAPture™ contact.

The Ø13 mm X 13 mm (1.6 cm³) $\text{LaCl}_3(\text{Ce})$ scintillation detector showed promise, with its combination of good detection sensitivity and moderate resolution; however, researchers planning to use these scintillators in measurement applications should carefully evaluate the impact of the unavoidable natural ^{138}La activity, and should inquire regarding the level of ^{227}Ac + D contamination in the crystal material to be purchased to avoid potentially serious application complications.

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