

# Study of Shadowing in Beta-Gamma Coincidence Plots for Radioxenon

# December 2018

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M P Foxe M W Cooper D A Haas J C Hayes J D Lowrey A M Prinke

Prepared for the U.S. Department of Energy under Contract DE-AC05-76RL01830

Pacific Northwest National Laboratory Richland, Washington 99352

# **Acronyms and Abbreviations**

GFE Government furnished equipment

HV High voltage

PMT Photomultiplier tube

PNNL Pacific Northwest National Laboratory

PXI PCI eXtention for Instrumentation

TBE Teledyne Brown Engineering

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# **1.0** Document Overview

This document describes a study of the cause of shadowing observed in certain beta-gamma plots produced with beta-gamma coincidence detectors for detection of radioxenon. Beta-gamma detectors must possess adequate resolution to allow for the metastable isotopes <sup>131m</sup>Xe and <sup>133m</sup>Xe to be distinguished from each other and from <sup>133</sup>Xe. One effect that can minimize the ability to distinguish the metastables from <sup>133</sup>Xe is the shadowing effect, in which a band of high-energy signal is present in addition to the primary signal. A study was performed to determine the cause of the shadowing, and how to minimize the effect of the shadowing on an individual beta cell.

Through this study, it is determined that the shadowing results from a difference in light transmission between the dome and end-cap portions of the beta cell. Consequently, it is not possible to remove the shadowing during the beta cell manufacturing process. The shadowing must be minimized by preferentially matching the light output from the end-cap and dome portion prior to the manufacturing process.

# 2.0 Beta-Gamma Plot Shadowing

#### 2.1 Beta Cell Construction

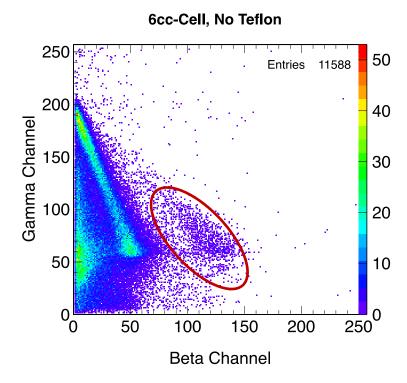
Plastic scintillator beta cells are commonly used in radioxenon detectors, **Figure 1**. The electrons interact within the beta cell producing a light signal proportional to the energy of the electrons. In certain instances, light is lost before it can be detected by the photomultiplier tube (PMT). Non-uniform loss of the light can result in the shadowing where there are two distinct signals for the same energy of electrons. This shadowing feature is demonstrated in **Figure 2**, and has the potential to both worsen the energy resolution of the meta-stable conversion electrons, and result in <sup>131m</sup>Xe counts to appear in the <sup>133m</sup>Xe ROI and <sup>133m</sup>Xe counts to shift out of the <sup>133m</sup>Xe ROI. These two effects result in an increased MDC on the xenon isomers.



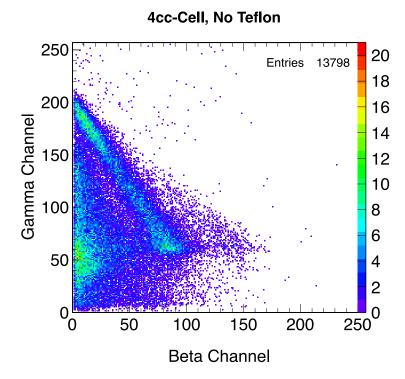
**Figure 1:** Schematic of the radioxenon detector, in which the beta cell sits inside the well of the NaI gamma detector.

# **2.1.1** Signal without any Teflon to show that it is an inherent issue

Teflon is used as a reflector for the scintillation light, directing a larger fraction of the light back into the beta-cell and to the PMT. Data was taken without the Teflon wrapping to verify if the shadowing effect is due to a spatial variation in the Teflon wrapping. Since the shadowing is observed when no Teflon wrapping is present, **Figure 2** and **Figure 3**, the shadowing is an intrinsic property of the beta cell, and not part of the post fabrication light reflector processing.



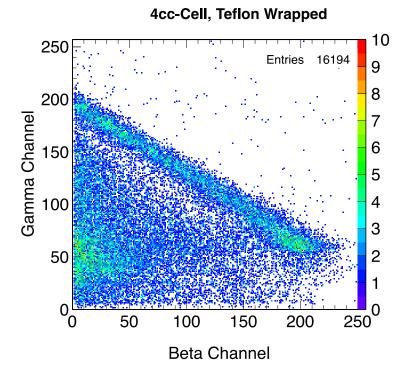
**Figure 2:** Beta-Gamma detector under the irradiation of a  $^{137}$ Cs source. The primary line has a beta endpoint of channel 50, while the shadow extends to ~125 (circled in red).



**Figure 3:** Beta-Gamma spectrum for a 4cc cell, with no Teflon wrapping. Since the shadowing appears without any Teflon wrapping, it is an intrinsic property of the cell and not one generated from the cell housing.

# 2.1.2 Effect of Teflon Wrapping

Wrapping with Teflon increases the light collection, shifting the primary signal line towards the shadow, **Figure 4**. This suggests that the shadow is due to near full light collection somewhere in the cell, and additional light is being lost somewhere to generate the primary signal line.



**Figure 4:** Beta-Gamma spectrum for a 4cc cell, with Teflon wrapping. The Teflon acts as a reflector, increasing the amount of light detected by the PMT. Since this removes the shadow in this instance, the shadow is a case of complete light collection, while the primary line is incomplete light collection throughout the bulk of the cell. Note that the Teflon does not always fix the shadow and is not a solution to the problem.

#### 2.1.2.1 Impact of Shadowing on MDCs

Shadowing has the potential to appear in beta-gamma spectrum, regardless of the efficiency of the reflector. Since shadowing appears in the complete setup of the detector, it can cause two problems: 1) the high energy shadow has the potential to worsen the beta resolution. 2) If the high-energy shadow is sufficiently high, the events from <sup>131m</sup>Xe can crossover into the <sup>133m</sup>Xe ROI, and the <sup>133m</sup>Xe events can extend out of the <sup>133m</sup>Xe ROI. If crossover occurs, then the MDCs for the meta-stables could increase significantly, as there would be additional interference ratios to be taken into account.

# **3.0** Beta Cell Parts and Signals

There are three primary parts to the beta cell, the scintillating dome, the scintillating end-cap, and the PMT for light collection. Examples of the dome and end-cap for the 6cc-wide, diffuse cell, and 6cc-long cell are shown in **Figure 5**. Once the end-cap and dome are connected, the cell is cemented to the PMT using a jig, which certifies that the cell is in the center of the PMT window, **Figure 6**. A completed 4cc cell, connected to a 1 inch PMT is shown in **Figure 7**. In an effort to determine which portions of the beta-gamma signal are due to each piece, we took a piece-by-piece testing approach.



**Figure 5:** The dome and end-cap portions of the 6cc-wide (left), diffuse (center), and 6cc-long (right) cells. The end-cap is a top-hat design for a more secure connection.



**Figure 6:** The jig used to center the beta cell on the PMT window for the gluing process. A weight is then placed on top of the jig to provide uniform pressure during the gluing process.



Figure 7: Completed 4cc cell glued to a 1-inch PMT.

# **3.1 PMT**

The PMT was tested individually to determine the signal due to any <sup>137</sup>Cs gamma rays interacting with the photocathode and producing secondary electrons that are amplified through the PMT dynode chain. The PMT shows no noticeable signal at higher energies and a very limited interaction rate due to the <sup>137</sup>Cs source. The <sup>137</sup>Cs source was placed adjacent to the PMT within the NaI well for increased gamma ray flux. The channel to energy ratio on **Figure 8** is 10x the ratio used with the beta cell connected so that signal could be better observed. The signal is all very low energy and nothing could produce the shadow.

## Bare\_PMT - Ch to Eng 10x Usual **Entries** Gamma Channel 250<sup>0</sup> Beta Channel

**Figure 8:** Beta-Gamma spectrum for the PMT under <sup>137</sup>Cs radiation. There is minimal interaction rate, and no signal above channel 150 in this figure, equivalent to channel 15 when a scintillator is attached. The vertical lines are a result of the increased gain to study the impact of the PMT.

## **3.2** Dome

Data was taken with the dome connected directly to the PMT using an optical pad. The primary line seen in the beta-gamma plots appears when just the dome is present, **Figure 9**, suggesting that dome is responsible for the lower light collection events.

## Diffuse Cell, With Teflon, No End-Cap **Entries** Gamma Channel 0, Beta Channel

**Figure 9:** Beta-gamma spectrum for the dome portion of a beta cell connected directly to a PMT using an optical pad.

# 3.3 End-Cap

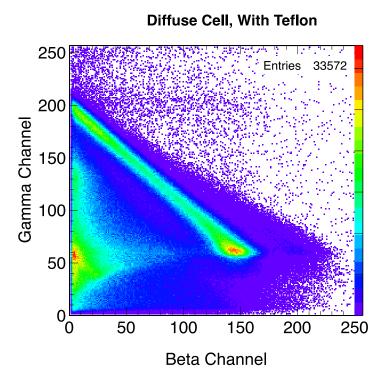
With only the end-cap connected to the PMT, the shadow is present without the primary signal suggesting that the shadow is due to increased light collection from events generated within the end-cap, **Figure 10**.

# Diffuse Cell, With Teflon, Only End-Cap 250 6546 Entries 200 Gamma Channel 150 100 50 0 50 100 150 200 250 Beta Channel

**Figure 10:** Beta-gamma spectrum for the end-cap portion of a beta cell connected directly to a PMT using an optical pad.

# 3.4 Both parts combined

When the dome and end-cap are both connected to the PMT, **Figure 9** and **Figure 10** are added together to give the results seen in **Figure 11**. With the shadowing present for the full cell, it suggests a material property and not a problem with the interface, but this was further investigated.



**Figure 11:** Beta-gamma spectrum for the dome and end-cap portions of a beta cell connected together and to a PMT using optical pads.

# 4.0 Optical Model of a Beta Cell

An optical model for a beta cell was generated to verify the potential loss of light collection from the dome portion of the beta cell. The light collection from a beta cell was modeled using a ray tracing program, which tracks the light propagation and final luminosity entering the PMT photocathode. Uniform air gaps of 0, 50, and 100 nm between the cell and the PMT were assumed for the model. The simulations shows that each 50 nm of air results in a loss of  $\sim$ 10% of the light generated within the cell, relative to the case of no air gap. This was noted to be of particular importance when a temporary construction method such as optical pads is utilized. The optical model was not used to quantitatively study the light loss, but focused on verifying the potential mechanisms.

Beta cells of volume 4 cc and 6 cc were simulated with the model. The increased length of the 6 cc cell results in an increased number of reflections and a lower light collection efficiency compared to the 4 cc cells. It could be possible that the dimensions of the 6 cc cell could be optimized to produce fewer reflections between the light generation and collection by the PMT, but that was not investigated in this study.

# **5.0** Beta Cell Investigation

Since optical pads were used in the previous tests, Section 3.0, there was uncertainty in the pressure applied to the cell-PMT interface. This uncertainty could result in an air-gap producing a shadow that would not be present in a fully fabricated cell. The previous generation of beta cells fabricated by Saint-Gobain and assembled by Matt Cooper and Justin McIntyre demonstrated minimal shadowing. A series of tests were performed to determine possible means of mitigating the degree of shadowing within the beta-gamma plots.

# **5.1** Reproduce Previous Cell Versions

#### 5.1.1 Excess Glue at PMT Interface

One investigated cause for the shadowing was the potential for a loss of light from the sides of the dome to end-cap interface. In the previous fabrication process, there was excess glue that could have potentially acted as a light guide into the PMT. In the new more precise fabrication process, there is minimal excess glue. In an effort to reproduce previous cell construction, excess glue was added to a cell with shadowing to try to increase the light collected from the dome portion of the cell. The PMT used for these tests was 31533, with dome 31533 and end-cap 31533 attached. A picture of the cell before and after the excess glue was added to the interface is shown in **Figure 12**. The results of the test, **Figure 13**, suggest that the amount of glue overflow outside of the cell-PMT interface has no noticeable effect on the degree of shadowing observed.

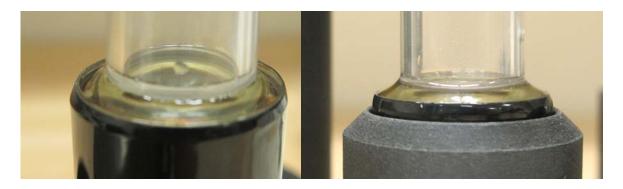
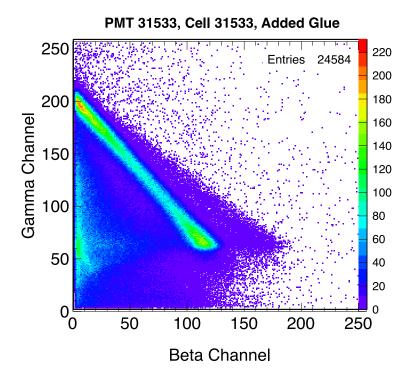
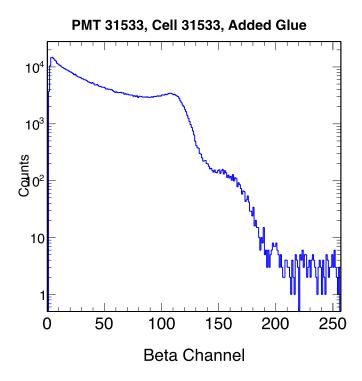


Figure 12: Cell connected to a PMT with minimal glue (left) and with the added excess glue (right).



**Figure 13:** Beta-gamma spectrum with excess glue added to the connection between the cell and the PMT.



**Figure 14:** Gated beta spectrum showing the second shoulder due to the shadowing, and no noticeable improvement from the excess glue.

# 5.1.2 Repeat Cementing Process

The optical model suggests that a uniform air gap of 50-100 nm could produce a decrease in light collection to account for the difference between the primary signal and the shadow. While unlikely, if there was a significant portion of the cell-PMT interface that included an air gap, it could produce the reduced primary signal, while the remaining area of the interface accounted for the shadow. To account for this possibility, an entire cell, which demonstrated shadowing, was removed from its respective PMT

and re-cemented to remove any air pockets that may have been previously present. The cell is shown before and after it was removed and re-cemented in **Figure 15**, with the results shown in **Figure 16**.



Figure 15: Cell used in the tests before (left) and after (right) the cell was removed and recemented.

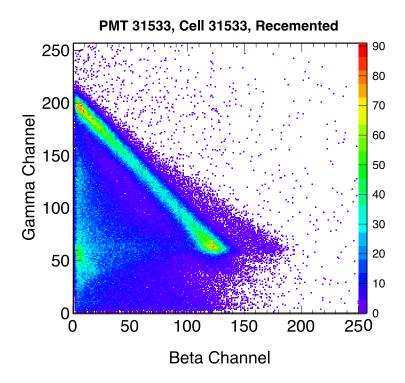
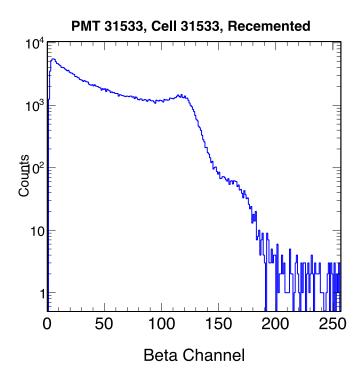


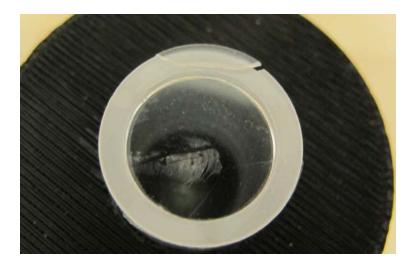
Figure 16: Beta-gamma spectrum for cell 31533 after it was removed and recemented to the PMT.



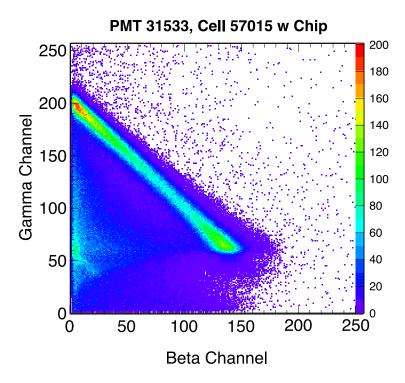
**Figure 17:** Gated beta spectrum after the recementing process, showing the second shoulder due to the shadowing.

# 5.1.3 Swap the PMTs used for Two Cells

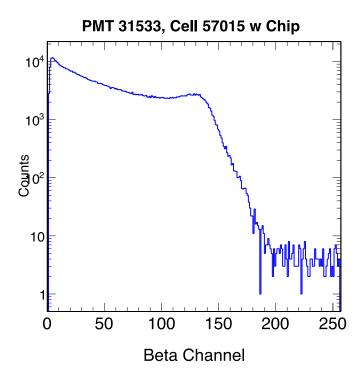
It is possible that the variation in light collection over the face of the PMT could produce a shadow like the one seen in the beta-gamma plot. Since the shadowing is seen as a digital variation, it would require the variation to be discrete as opposed to a continuous function. By swapping the PMT of a cell that doesn't demonstrate shadowing with one that does show shadowing, it allowed for the cause of the shadowing to be attributed to either the PMT or the cell. The cells and PMTs used in these tests were the good cell/PMT with no shadowing, 57015, along with the bad cell and PMT combo, 31533. Since no change in the degree of shadowing occurred, it eliminated the PMT as a cause of the shadowing. As a control test, PMT/cell 57894 went through all of the tests but kept the same configuration. With no change in the control cell, decreases the likelihood that it is a fabrication procedure that creates the shadowing feature. During the deconstruction process, a small piece of the 57015 end-cap broke off producing a chip in future tests.



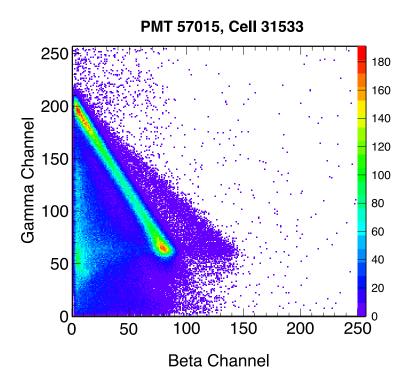
**Figure 18:** End-cap of cell 57015 showing the chip from the end-cap that occurred during separation. This was cemented back in place for remaining tests.



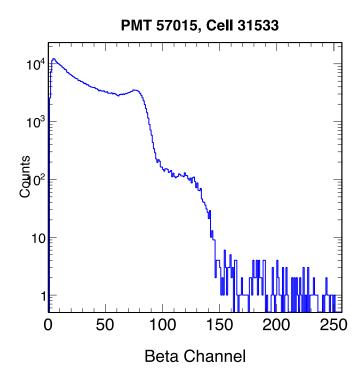
**Figure 19:** Beta-gamma spectrum for cell 57015 (good) on PMT 31533 (bad), showing no noticeable shadowing.



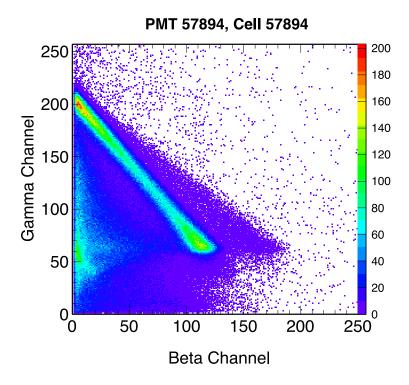
**Figure 20:** Gated beta spectrum for cell 57015 (good) on PMT 31533 (bad), showing no noticeable shadowing.



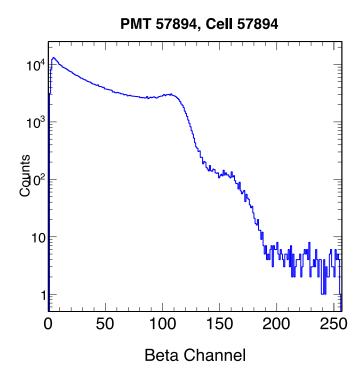
**Figure 21:** Beta-gamma spectrum for cell 31533 (bad) on PMT 57015 (good), showing no improvement to the shadowing due to the good PMT.



**Figure 22:** Gated beta spectrum for cell 31533 (good) on PMT 57015 (bad), showing no improvement to the shadowing due to the good PMT.



**Figure 23:** Beta-gamma spectrum for cell 57894 (bad) on PMT 57894 (bad), showing the same shadowing as the previous tests.



**Figure 24:** Gated beta spectrum for cell 57894 (bad) on PMT 57894 (bad), showing the same shadowing as the previous tests.

#### 5.1.4 Swap the Dome and End-Cap for Two Cells

Instead of swapping an entire cell, the dome from a good cell was switched with that of a bad cell. This allowed for the testing of two distinct causes for the shadowing effect. If the loss of light is due to the interface of the dome and end-cap, then the shadow could potentially follow the dome to the new end-cap/PMT setup, **Figure 25** - **Figure 30**. If the shadowing effect is due to the end-cap being much more efficient at producing light detected by the PMT, then the shadow would stay with the end-cap/PMT. When new cells are built, in order to minimize the chance of observing the shadowing effect, the dome and end-cap can be tested individually, to gain match the light production and collection. After switching the end-caps for the good cell (57015) and the bad cell (31533), it was concluded the shadow is due to a variation in the light production of the dome and end-cap. This variation could be due to both a decreased

light production in the dome, Figure 31 and Figure 32, as well as an increase light production in the endcap, Figure 33 and Figure 34.

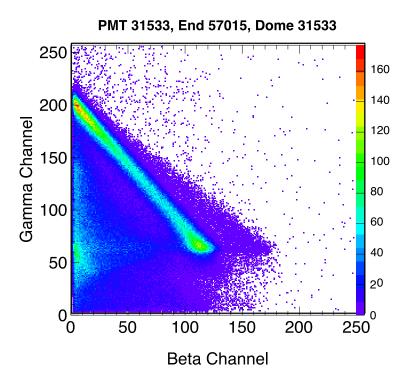


Figure 25: Beta-gamma spectrum for PMT 31533, end-cap 57015, and dome 31533.

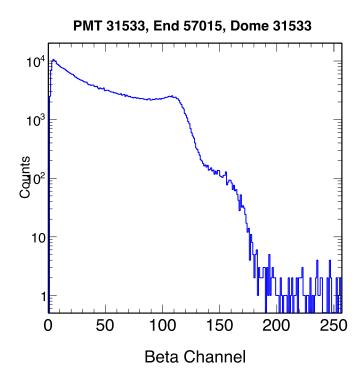


Figure 26: Gated beta spectrum for PMT 31533, end-cap 57015, and dome 31533.

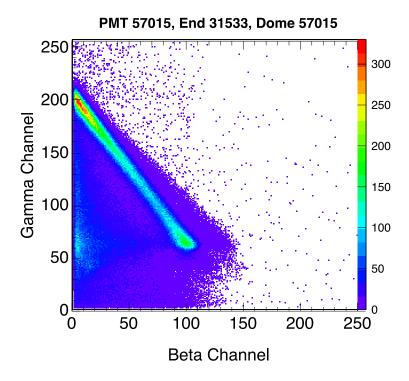


Figure 27: Beta-gamma spectrum with PMT 57015, end-cap 31533, and dome 57015.

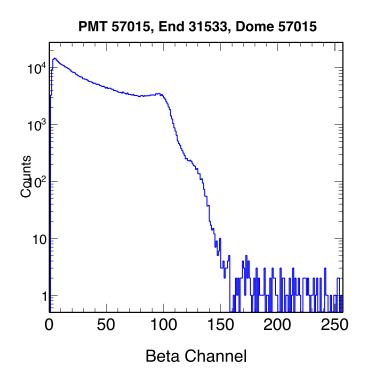


Figure 28: Gated beta spectrum with PMT 57015, end-cap 31533, and dome 57015.

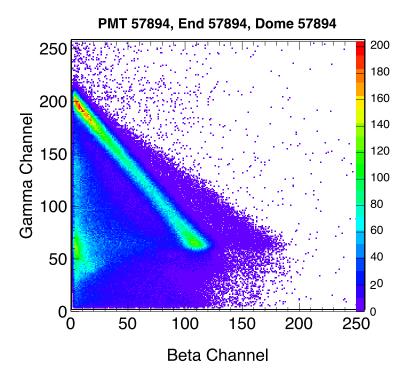


Figure 29: Beta-gamma spectrum with PMT 57894, end-cap 57894, and dome 57894.

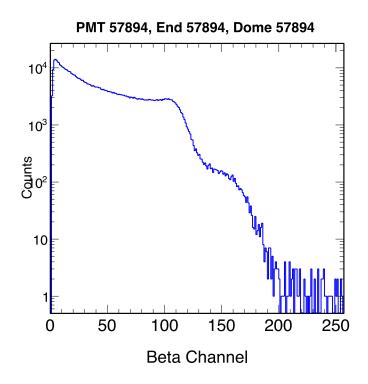
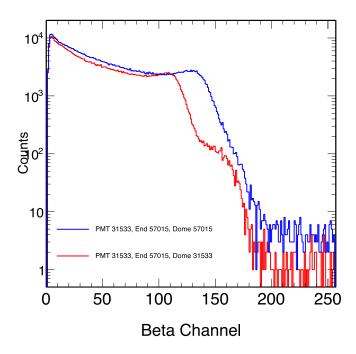
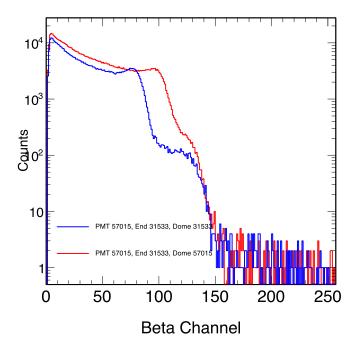


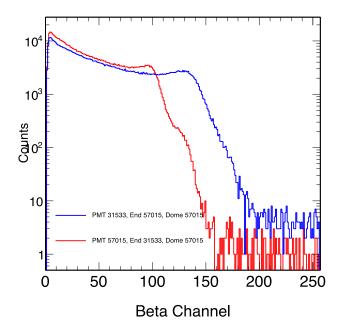
Figure 30: Gated beta spectrum with PMT 57894, end-cap 57894, and dome 57894.



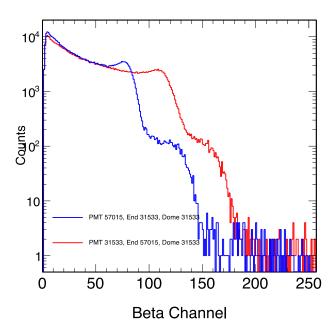
**Figure 31:** Gamma-gated beta spectrum for a cell showing little shadowing (blue), and with the dome of a cell which shows significant shadowing (red). The shadowing effect follows the dome, suggesting a material property.



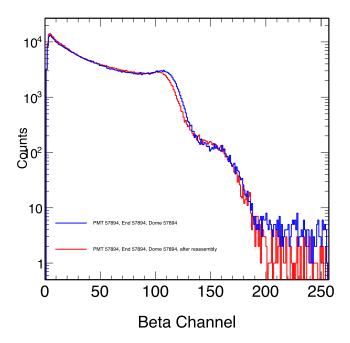
**Figure 32:** Gamma-gated beta spectrum for a cell showing significant shadowing (blue), and with the dome of a cell which shows little shadowing (red). The shadowing effect decreases with the dome of a good cell, suggesting a material property.



**Figure 33:** Gamma-gated beta spectrum for a cell showing little shadowing (blue), and with the end-cap of a cell which shows more significant shadowing (red). The red end-cap produces more light than that of the blue, contributing to the shadowing effect. Note that the end-point of the red curve is decreased due to a different PMT being used for the measurement.



**Figure 34:** Gamma-gated beta spectrum for a cell showing significant shadowing (blue), and with the end-cap of a cell which shows slight shadowing (red). The red end-cap produces less light than that of the blue, decreasing the shadowing effect. Note that the end-point of the blue curve is decreased due to a different PMT being used for the measurement.



**Figure 35:** Gamma-gated beta spectrum for a cell showing significant shadowing before deconstruction (blue), and after reassembly (red). The reproduction of the same plot serves as a control to show that the changes in **Figure 31** - **Figure 34** are due to the end-cap/dome switches.

# **5.2** Unpolished Cell Fabrication

The dome and the end-cap have previously been fabricated from two different batches of BC-404. The end-cap was diamond milled with an optical smoothness of 0.2 microns, while the dome went through a polishing procedure and had an optical smoothness of 1-2 microns. A new batch of cells was obtained from the manufacturer using a single batch of BC-404 and machining everything to an optical smoothness of 4-5 microns without using any polishing procedure to avoid any possible light production contaminants.

Through similar tests as in the previous sections, it was observed that the difference in polishing procedure did not impact the degree of shadowing present. It is possible that the way the end-caps are manufactured is different from that of the domes, resulting in a difference in light collection.

# **6.0** Shadowing Mitigation

Since one primary cause for the shadowing effect is a mismatch in light production/collection between the dome and the end-cap, new cells should be characterized and the parts should be selected in an effort to minimize the number of completed cells that show shadowing. The following testing program is designed to aid in the production of cells, which exhibit reproducible uniform light production and collection.

#### **6.1** New Cell Test Procedure

# 6.1.1 Serial Number System

Each part should be stored in a bag with the part number labeled on the outside. This should be kept track of for the PMT, PMT base, cell end-cap, and cell dome. This will produce a chain-of-custody so that any issues can be tracked back to the source and so the cells can be properly gain matched.

# 6.1.2 Test Cell Components

Cell components should be tested for shadowing before manufacturing or memory effect coatings. This will allow for the light collection of the dome and end-cap to be measured, along with a preliminary measurement of the energy resolution of the beta cell. If it is decided to move forward without a solution to the shadowing problem, it is desirable to measure the light collection for each component and gain match the domes with the end-caps. This would result in a ~50% loss of cells, but could result in a higher quality final product. This is also something that could be designated to the plastic manufacturer in order to gain match the dome and end-cap prior to shipment.

#### 6.2 Saint-Gobain Beta Cells

Two batches of Saint-Gobain cells were ordered and tested. The first batch of 6 cells had 4 cells with no shadowing observed and two cells with minimal shadowing present. With the positive outlook for the Saint-Gobain cells, a second batch of 12 cells was ordered. The second batch of cells was tested with the same procedure used in the testing of new cells. Unfortunately, the second batch of cells showed the same 50:50 fraction that was observed with the Eljen cells, suggesting that the batch of 6 cells showed a false sense of improvement due to low statistics.

#### **7.0** Conclusions

Throughout this study, it was established that the shadowing is no a by-product of the fabrication process. The shadowing results in a difference between the light collection on the event-by-event level. One leading candidate for this difference in signal appears to be an effect of the difference in light transmission from the two portions of the plastic scintillator beta cell. It should be possible to engineer the plastic scintillator pieces with a smaller difference in light transmission, but this may require a new research project. Additionally, the end-cap could be made out of non-scintillating material to remove the shadowing completely, but this would result in a decrease of ~10% in the efficiency of the beta cell. It is desirable to perform light transmission matching between the dome and end-caps in a batch of beta cell parts. If this matching process is performed prior to the fabrication of the beta cells, the reduction in shadowing is expected to be a long term effect for the life of the beta cell. Additional data will be required if the shadowing effect for a beta cell is determined to be time dependent.

A second candidate for the difference in signal is an electrical artifact in the data acquisition system. It was determined that in some cases the shadowing could be mitigated by varying the settings on the Pixie-4 signal processing, though it is not clear if this applies to all of the instances of shadowing. In that specific instance, the shadow was enhanced due to a reflection in the beta signal between the cell and the Pixie-4 input, caused by the impedance mismatch we deliberately introduce for timing reasons. The beta signal reflection was big enough and far enough delayed from the main beta signal to get counted as a

separate, smaller event; it was close enough in time to get counted as a coincidence at the same gamma energy. Changing the signal processing properties (time gates or thresholds) changed whether there was a shadow. The cable length would also have a potential effect due to changes in reflection time and cable impedance.

The combination of these tests shows that the shadowing could be present due to both physical characteristics and electrical settings. There is no perfect method for removing the shadowing from all beta cells, but it can be reduced through specific fabrication and setup techniques.



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