Air-Transfer Production Method for Large-Area Picosecond Photodetectors

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We have designed and prototyped the process steps for the batch production of large-area micro-channel-plate photomultipliers (MCP-PMT) using the "air-transfer" assembly process developed with single $LAPPD^{TM}$ modules. Results are presented addressing the challenges of designing a robust package that can transmit large numbers of electrical signals for pad or strip readout from inside the vacuum tube, and of hermetically sealing the large-perimeter window-body interface. We have also synthesized a photocathode in a large-area low-aspect-ratio volume, and have shown that the micro-channel plates recover their functionality after cathode synthesis. These steps inform a design for a multi-module batch facility employing dual nested low-vacuum (LV) and ultra-high-vacuum (UHV) systems in a small-footprint. The facility design provides full access to multiple MCP-PMT modules prior to hermetic pinch-off for leak-checking and real-time photocathode optimization.

I. INTRODUCTION

Applications for large-area coverage of photodetectors with excellent time and space resolution include: a) correlated precision time and space information in high-energy particle collider and fixed-target events¹; b) simultaneous measurements in near and far detectors of neutrino oscillations using time-selected energy spectra²; c) imaging double-beta decay events in large liquid scintillator detectors using timing to separate Cherenkov and scintillation light^{3–8}; d) reconstruction of neutral mesons decaying to photons in searches for rare meson decays^{9–11}; e) low-dose whole body Time-of-Flight Positron-Emission Tomography^{12–15}; and e) nuclear non-proliferation and reactor monitoring¹⁶.

The typical method of production of commercial photomultipliers (PMT) is in a batch production station that processes multiple modules per cycle. Figure 1 shows a commercial PMT station¹⁷ purchased by the LAPPD Collaboration¹⁸. The PMT process steps served as the model for the development of the batch production process for large-area flat-panel micro-channel-plate-based (MCP-based) photodetectors (MCP-PMT) ^{19,20}.

The salient features that form the basis of the design for flat-panel photodetector production are as follows¹⁷. During processing the PMT glass envelopes are at ultra-high vacuum (UHV) internally and are at atmosphere externally. The initial evacuation of the phototube is at low vacuum (LV), performed by an external pump through long low-conductance plumbing, with the PMT glass envelopes connected through a narrow tubulation sealed by a low-vacuum O-ring gland. An oven is placed over the assembly for bake-out and subsequent photocathode formation. The antimony pre-cursor layer is evaporated by line-of-sight onto the inside surface of the entrance window, and the alkali vapor is generated by sources inside the glass envelope. The UHV inside the tube is provided by

the bake-out, the internal getter, and the alkali vapors, and not by the external pump. After cathode optimization the tubulation is pinched off hermetically sealing the phototube with the getter inside.



FIG. 1. The photomultiplier batch production station purchased from the Photonis Lancaster plant¹⁷ that served as the model for development of the LAPPD batch production process.

Experience with the process steps used for the Photonis PMT station guided the development of the process for large-area micro-channel-plate-based PMT batch production described below. However, large-area MCP-PMTs present unique challenges, requiring: 1) a robust package that can

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transmit large numbers of electrical signals for pad or strip readout from inside the vacuum volume; 2) hermetically sealing the large-circumference window-body interface; 3) synthesizing the photocathode in a large flat package; and 4) recovery of the functionality of the large surface-area MCPs after cathode synthesis.

The synthesis of bialkali photocathodes by exposing an antimony (Sb) layer pre-deposited on a substrate by thermal evaporation was described by Sinclair in 2009²¹. This suggestion led to a DOE-funded development program for a theorybased *K*₂*CsSb* cathode in a collaboration of the University of Chicago (UC), Brookhaven National Laboratory (BNL) and Radiation Monitoring Devices (RMD, Inc.)^{22–25}. However, we recently discovered that the chemistry of photocathode synthesis based on a pre-deposited Sb layer had been explored in detail by Barois et al. in 1989^{26,27}, and that the air-transfer process is currently being used commercially in Russia to make multi-anode photomultipliers²⁸. Hamamatsu also processes some of their phototubes using the air-transfer process²⁹.

The Barois insitu photocathode process tightly controls the uniformity of the cathode layer through the thickness of the Sb precursor layer, which can be made highly uniform by conventional thermal evaporation³⁰. The chemical and physical composition of the structure is determined by a number of parameters as discussed in Refs^{22–25}, and has the attractive property that the reaction runs to completion.

Specific Cs-Sb compounds may be formed by equilibrium reaction of Sb with Cs vapor²⁷, the stoichiometry set by temperatures and pressures in the system. Some stoichiometries are optimal for high quantum efficiency. Since the Sb is immobile, a cathode of uniform thickness and composition can be produced from an Sb layer of uniform thickness and crystal structure^{24,25,30}.

II. THE DUAL-VACUUM AIR-TRANSFER METHOD

The LV/UHV-dual-vacuum technique uses an outer vacuum vessel which contains the photodetector module. The outer vessel provides thermal insulation for a high-temperature bakeout of the photodetector module ("tile"), reduces the formation of oxides in the solder during heating, and ensures that no mechanical pressure differential exists across the molten seal. The module is assembled with the entrance window clamped in place, and is connected through a tubulation or tubulations to an independent UHV manifold and pump. Initially, before the hermetic seal is formed during the bakeout thermal cycle, the two vacuum systems can communicate through the unsealed seam between the window and the tile base. The bake-out cycle forms the seal between the window and base, and activates the getter in the module. After cooling below the seal melting temperature the two vacuum systems are independent, and the outer vacuum can be opened to atmosphere. The inner vacuum system is used as a source for the alkali vapors to synthesize the photocathode from the Sb precursor layer on the window.

Figure 2 shows the outer vacuum vessel. The vacuum pump

for the outer system is on the left; the connections to the inner vacuum system enter through the flange on the bottom. The upper part of the vessel can be lifted off after sealing, exposing the sealed module for leak-checking and subsequent photocathode synthesis.

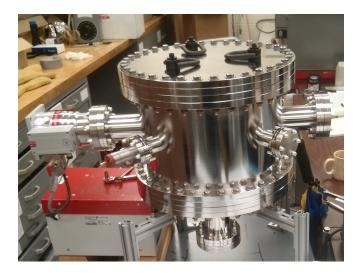


FIG. 2. The outer LV vacuum vessel that provides thermal insulation to the photodetector module during bake-out, a low oxidation environment, and equalization of the pressure across the molten indium alloy seal.

The inner UHV vacuum system consists of the tile volume, copper tubulations, and the manifold that connects the tile to the inner system vacuum pump and the alkali vapor source. A diagram of the manifold and the associated component process temperatures during alkali transport is shown in Figure 3.

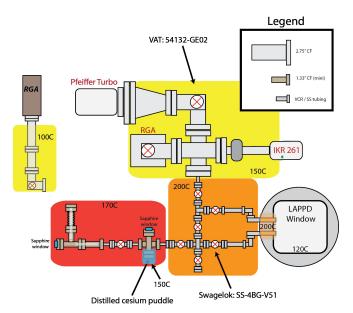


FIG. 3. The physical layout and associated component process temperatures during alkali vapor transport of the UHV inner vacuum manifold.

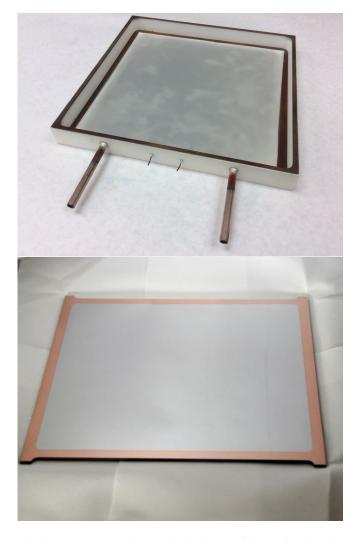


FIG. 4. Top: A two-pin ceramic tile body with metalized sealing surface and NiCr capacitively-coupled anode; Bottom: An entrance window with metalized sealing surface.

III. THE LAPPDTM PACKAGE

The UC ceramic tile package consists of a monolithic ceramic tile base with capacitively coupled anode and a transparent entrance window, shown in Figure 4. The fast signal pulses are capacitively coupled through the anode on the bottom plate³¹, avoiding the difficulty in brazing a large number of pins.

The tile base is one-piece, composed of high-purity Al_2O_3 ceramic^{32,33}. Two 1/4" OFHC copper tubes are brazed through the sidewall to provide for pump-down and alkali vapor transport. Current designs incorporate 6 copper pins, 3 per side, brazed through opposing sidewalls for HV distribution to the cathode, amplification section, and anode³³. Figure 5 shows a ceramic sidewall with pins and tubes.

The top surface of the tile base sidewall is the surface that mates with the window to provide the hermetic seal. The solder seal requires that this surface be metalized. The majority of the trials used thin film evaporation to deposit 200 nm

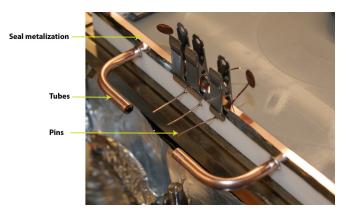


FIG. 5. Copper tubes that connect the tile base to the UHV manifold, and copper pins that provide HV to the cathode and amplification stack.

of 80-20 NiCr followed by 200 nm of Cu without a vacuum break³³ on the sealing surface, with the sidewall and anode tile surfaces protected by masks.

The bottom interior surface of the tile was also coated by evaporation. The first step was evaporation of 10 nm of NiCr on the active area of the anode to provide capacitive coupling to the external signal board³¹. A second set of masks was used to deposit 200 nm of NiCr followed by 200 nm of Cu, also without a vacuum break, for a narrow border overlapping the 10 nm NiCr anode to provide electrical contact³³.

The UHV inside the module is maintained after sealing by an internal non-evaporative getter consisting of two stacks each containing six 174-mm by 8-mm strips of SAES ST707³⁴ held in clips soldered to the anode.

The mating surface of the window was also metalized by evaporation³³ using successive masks³⁵. First 200 nm of NiCr was deposited on the border with thin 'fingers' toward the center to distribute current across the cathode. A second evaporation deposited 200 nm of Cu on the Ni border for the indium alloy solder bond. A third evaporation deposited a thin Sb film as the pre-cursor for the alkali cathode.

The unsupported spans of the window and tile base anode surface need to support almost 1000 lbs of compressive atmospheric pressure. Sixteen columns of 3/8"-diameter ceramic cylinders ("buttons") are placed internally at 1.8" spacing in the gaps between the window, MCP's, and anode. The buttons serve three purposes:

- Supporting the package against the compressive force of atmospheric pressure on the top and bottom of the package;
- 2. Setting the gaps between the MCP's, tile base, and the photocathode;
- 3. Applying mechanical force between the MCP surfaces and the high-voltage internal electrodes that connect the accelerating high voltages to their respective pins through the sidewall.

The left-hand panel of Figure 6 shows the placement of the button columns and the internal high-voltage electrodes connected to the side-wall pins during the first stage of assembly.

Atmospheric pressure is used to constrain the amplification stack against lateral motion; the right-hand panel shows the predicted stress for a 2.75 mm-thick B33 glass window.

The amplification section consists of two ALD-coated 20 cm-square MCPs in a chevron configuration^{36–38}. The spacing between plates is set by button spacers to 0.080"; the spacing between the top or bottom plate and the top (cathode) or bottom (anode) surfaces, respectively, is similarly set to 0.25". The capillary seal (see Section IV) reduces the necessary assembly tolerances by precisely determining the indium layer thickness via a 'hard-stop' in the seal gap rather than by the stack height. The measured precision on the column heights is 0.5 mils. Before assembling the tile the micro-channel plates were vacuum-baked and scrubbed with a UV lamp.

The monolithic ceramic tile avoids the problem of having many penetrations to bring the signals out by capacitively coupling through the thin metal anode to an external signal pickup board³¹. The signal board is customized to use microstrips^{39,40} or pads³¹, determined by the application.

IV. THE HERMETIC WINDOW INDIUM-ALLOY SEAL

The hermetic seal is one of the challenges of scaling up MCP-PMTs in size. A solution consists of clamping the window in place on the tile base at a set gap width, typically 0.002", and 'wicking' molten indium alloy⁴¹ into the gap by capillary action from wire placed contiguous to the gap⁴². The sealing surfaces require metalization prior to assembly. Both sealing surfaces are coated by evaporation with 200 nm of NiCr followed by 200 nm of Cu.

The tile base, inner amplification components, and window are clamped in place in a mechanical 'fixture' prior to the thermal sealing cycle. The fixture consists of a 0.50"-thick SS bottom plate that supports the tile base, a compression fixture to clamp the window onto the base from above, heating elements, and thermocouples for measuring the temperature during the process cycle. The copper tubulations are connected to the manifold to complete the inner vacuum system. Figure 7 shows an assembled tile in the fixture prior to sealing.

To bake out the tile and form the hermetic seal, two heating units, shown in Figure 7, consisting of 3mm-thick 6"-square ceramic plates wound with NiCr heating ribbon⁴³ are placed in thermal contact with the lower and and upper compression plates, as shown in Figure 7. The outer vacuum vessel is not heated.

Figure 8 shows the thermal cycle for Tile 31. The cycle consists of a ramp up of approximately 15 hours, a bake-out of 18 hours at 290 $^o\mathrm{C}$, followed by cool-down to room temperature. The feature around 220 $^o\mathrm{C}$ is due to the formation of the indium alloy capillary seal.

Leak checking after sealing is performed in two steps. A global check is performed before opening the outer vacuum vessel using a Residual Gas Analyzer (RGA)⁴⁴ by back-filling the vessel with argon with the tile isolated from the UHV (inner) manifold. After several hours, the pump to the manifold is valved off, and the valve between the manifold and the tile is re-opened. The RGA can then detect through the manifold

any argon accumulated in the tile. We estimate the sensitivity of the global check to be $1-2 \ 10^{-13}$ mbar-liters/sec.

If a leak is detected, after the outer vessel is opened a systematic local search is performed with a helium leak detector 45 attached to the manifold and a needle probe helium source. We estimate the sensitivity of this local check to be several 10^{-11} mbar-liters/sec.

V. BAROIS PHOTO-CATHODE SYNTHESIS

The air-transfer assembly process involves depositing a Sb pre-cursor layer on the window prior to assembly. After the sealing cycle is complete and the tile is leak-tested, the tile is re-heated to a temperature of $\approx\!120~^{o}\mathrm{C}$, below the melting point of the solder seal, for photocathode synthesis by introducing alkali vapor^26,27.

The bialkali K_2CsSb photocathode cathode compound was synthesized directly as part of this program^{22,23,46}. In the process development described here only a single alkali, Cs, was used for simplicity.

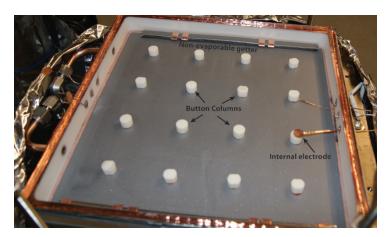
The cesium vapor source is shown schematically in Figure 9. A glass ampoule of $Cs^{47,48}$ is inserted in the vertical bellows section of the tee in the left-hand (ampoule) chamber. The ampoule is broken from above with a ceramic rod by compressing the bellows, as shown in Figure 9. The Cs liquid and glass ampoule can be observed through a sapphire window. The liquid Cs is then evaporated away from the glass shards of the ampoule into the right-hand (source) chamber by heating the ampoule chamber 49 . The valve between the source chamber and the ampoule chamber is then closed. A sapphire window in the source chamber allows visual monitoring of the surface of the distilled Cs. Cesiation of the tile is initiated by opening the valve to the manifold with the source chamber at 150-155 o C.

A 2-dimensional motion stage for mapping the photocathode response during photocathode synthesis is shown in Figure 10. The module is still connected to the UHV manifold; pinch-off occurs only after leak-checking and cathode characterization.

To measure the photo-response we applied a 37 V bias to the gap between the photocathode and the top surface of the first MCP. A 405 nm light-emitting diode was pulsed in phase with a lock-in amplifier and pico-ammeter to measure the current.

Figure 11 shows successive maps of the photo-response during the activation of the pre-cursor Sb layer by Cs vapor. The response grows to a saturation value near the inlet and then continues to the far side of the 8.26"-square window. The response saturates near 3% quantum efficiency (QE).

Neither the uniformity nor the end-point QE are satisfactory for a useable commercial device. We attribute the lower quantum efficiency near the edges to a non-uniform temperature of the window during synthesis. The tile was heated through its contact with the fixture lower plate, and was subject to convective cooling from above. Thermally isolating the assembly from convection should eliminate the non-uniform temperature of the window. In addition, the QE saturated, but



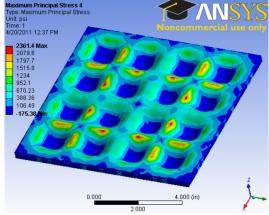


FIG. 6. Left:The tile base during assembly showing the spacer buttons and their connecting internal high-voltage electrodes. At the top of the picture the getter is positioned to be soldered to the NiCr anode layer during the bake-out thermal cycle. Right: A finite-element analysis of the stress on a B33 window under vacuum.

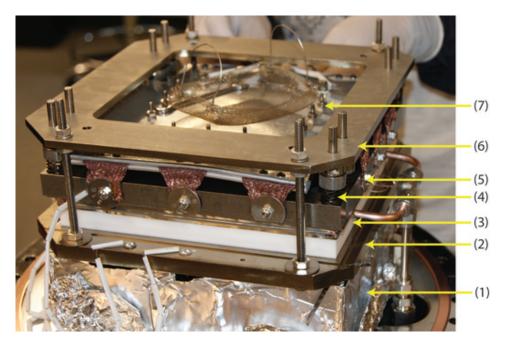


FIG. 7. An assembled tile clamped in place in the compression fixture prior to sealing. Heaters above and below the tile couple to the top and bottom fixture plates. The indicated components are: (1) the lower NiCr heater assembly; (2) the bottom compression fixture plate; (3) the tile assembly; (4) the compression mechanism; (5) floating rigid press bars; (6) the upper compression fixture plate; and (7) the upper NiCr heater assembly.

at a lower value than is attainable for a good Cs_3Sb cathode⁵⁰. We attribute this to the overly-thick Sb precursor layer. The overall Cs vapor migration time through the tile in Fig.11 can be decreased by increasing the processing temperature.

Figure 12 shows that the micro-channel plates recover functionality after cesiation. There is an initial large drop in resistance that would preclude operation. After the alkali vapor is valved off, the plates increase in resistance, allowing operation after cooling with increased gain, but with high noise. The gain and noise then fall to operational values within a few hours.

After photocathode synthesis and characterization, the cop-

per tubulations are hermetically sealed by pinching off⁵¹. The razor-sharp edge of the pinch is then potted for safety, and the tile can be removed from the fixture.

VI. CONCLUSIONS

Large-area MCP-based photomultipliers employing ALD coated plates have unique performance parameters, with typical gains greater than 10⁷, amplification-section (as opposed to photocathode) noise less than a few Hz, sub-mm resolutions in each transverse spatial dimension, and time resolution

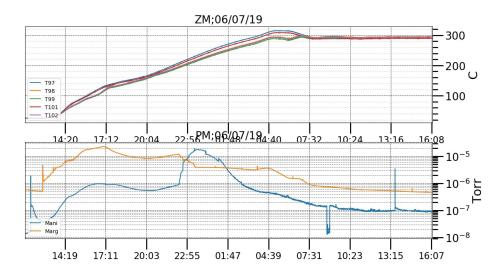


FIG. 8. The thermal cycle temperature and dual vacuum pressure data from Tile 31. The feature around 220 o C is due to the formation of the capillary seal between the two vacuum systems after the In-Ag alloy has melted at a temperature around 143 o C.

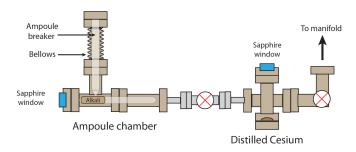


FIG. 9. Diagram of the alkali (Cesium) vapor source. The left-hand section is the ampoule chamber; the right-hand section is the source chamber.

of 5-10 psec for charged particles and 30-50 psec for single photons^{40,52}. The small feature size, concomitant good resolutions, and high gain are intrinsic rather than highly tuned⁹. The ALD-based functionalization of the glass capillary substrates^{36,37} eliminates the shortened tube lifetime due to ion feedback^{1,53}.

At the outset of this work the major obstacles to PMT-like batch production of large-area MCP-PMTs were identified as: a) the long and rectangular hermetic seal; b) 'in-situ' synthesis of alkali photo-cathodes in a large-area low-profile package; c) compromised performance of the MCPs after exposure to alkali vapors; and d) a robust package that supports large numbers of high bandwidth electrical signal connections to an external readout system. We address these in turn below.

The demonstrated solution to the hermetic seal relies on: a) clamping the window at the pre-set width of the solder seal layer prior to heating; clean Au or Cu surfaces on the window and base with no solder material (and hence no oxides) in the gap prior to sealing; b) allowing capillary action to 'wick' the molten alloy into the window-base gap during bakeout away from oxide; and c) use of In-Ag alloy as solder.

After this work started, we discovered that the chemistry

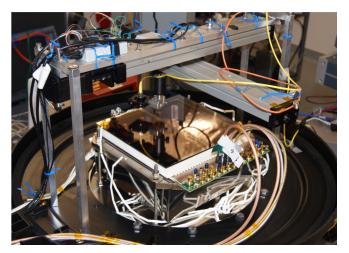


FIG. 10. A 2D scanning stage for mapping photo-response during and after photocathode synthesis.

of the 'in-situ' photo-cathode synthesis starting with a predeposited uniform thickness Sb layer was studied in depth 30 years ago^{26,27}. We also discovered that the air transfer of the window with a pre-deposited Sb layer is currently in use commercially²⁸. We note that Ref.²⁷ states "Manufacturing photo-emissive layers by equilibrating the antimonides with binary alkali vapors would lead to a product with a better definition than those obtained through the dynamic process used at present".

The recovery of MgO-coated MCP functionality after exposure to Cs vapor during photocathode synthesis was demonstrated. The plates initially drop dramatically in resistance, and both gain and noise increased, but recover with subsequent operation before pinch-off.

Lastly, the development of capacitive coupling of an internal thin metal anode layer to an external, application-specific, signal-pickup printed circuit board enables the decoupling of

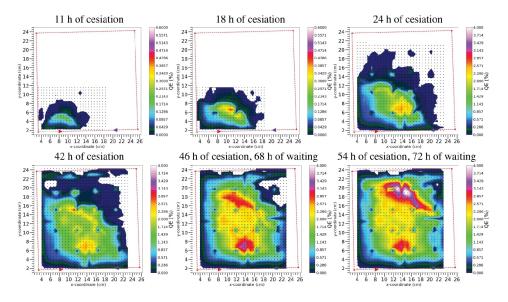


FIG. 11. Successive maps of the photo-response for Tile 31 during photocathode synthesis.

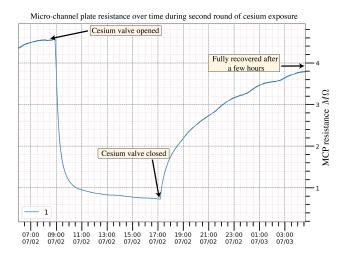


FIG. 12. The resistance of the stack of two microchannel plates and spacers during a cesiation cycle, showing that the plates recover functionality after an initial large drop in resistance.

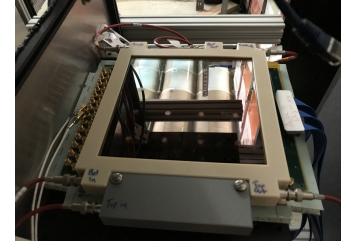


FIG. 13. The UC LAPPD TM Tile 31 with a signal readout board and HV connections.

the photomodule design from the optimization of pads or strips for high-bandwidth signals for applications requiring many pads or strips in the readout³¹.

We have shown that each of these potential 'show-stopper' obstacles is technical rather than fundamental. A proof-of principle solution to each that is scalable to batch production is described above. However, the development of an optimized industrial batch process for bringing volume up and cost down for wide-spread use in particle physics, medical imaging, and nuclear security remains as the task for commercialization.

VII. ACKNOWLEDGEMENTS

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The data that support the findings of this study are available from the corresponding author upon reasonable request.

The authors hold three patents related to this work 19,20,42.

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