Light Readout Optimisation using Wavelength Shifter - Reflector Combinations

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Abstract. The use of reflectors coated with a wavelength shifter (WLS) along with standard bialkali PMTs is an economical method for an efficient readout system for vacuum ultra violet (VUV) light produced in large liquid argon detectors. Various thicknesses of tetraphenyl butadiene (TPB) were deposited by spraying and vacuum evaporation onto both specular 3M™-foil and diffuse Tetratex™ (TTX) reflectors. 128 nm VUV light generated in 1 bar argon gas by a 5.4 MeV α source was detected by a 3-inch bialkali borosilicate PMT within a 1 m tube lined internally with a TPB coated reflector. The light collection was recorded as a function of separation between source and PMT for each combination of coating and reflector for distances up to 1 m. Reflection coefficients of TPB coated reflectors were measured using a spectroradiometer. WLS coating on the PMT window was also studied. The optimum coating and reflector combination was TPB evaporated on TTX. Measurements with coating thicknesses of 0.2 mg/cm² and 1.0 mg/cm² yielded a similar performance. The best PMT window coating is obtained by TPB evaporation of 0.05 mg/cm².

1. Introduction
Tonne scale liquid argon (LAr) targets such as that used in the Argon Dark Matter (ArDM) experiment [1, 2, 3] typically require in excess of ten large area PMTs for acceptable light readout. The argon scintillation light due to neutral or charged particle excitation is in the vacuum ultraviolet (VUV) centered at about 128 nm [4, 5, 6]. Currently large MgF₂ windowed PMTs that can detect 128 nm light are commercially not available. An alternative technique is to apply wavelength shifting chemicals on borosilicate windowed PMTs thus shifting VUV argon scintillation into the visible spectrum, the typical quantum efficiency (QE) of a cryogenic (Pt-underlay) borosilicate windowed PMT being from 10 to 15% at approximately 430 nm. Tetraphenyl butadiene (TPB) powder has been used which has an above average Stokes shift and can absorb 128 nm light, emitting in the required visible PMT region [7, 8, 9, 10]. The argon scintillation light is characterised by two distinct decay times - a slow component, \( \tau_2 \) (triplet eximer), and a fast component, \( \tau_1 \) (single eximer)[5, 11]. Determination of the time constants is through a multiple parameters nonlinear least square fit with additional degrees of freedom related to the height, start time and baseline of the pulses. The decay time of the slow component, \( \tau_2 \), increases with the increase of argon purity and therefore can be used as a measure of the purity of argon. From the literature the purest gas argon has a \( \tau_2 \) of 3200 ± 300 ns [11]. The analysis of the measurements described in subsections 3.2, 3.3 was based around this property of argon scintillation. Section 2 describes TPB coating methods and sample
preparations. Section 3 outlines three experiments aiming for reflector selection, optimisation of TPB coating thickness and deposition technique and presents the results obtained. These results led to the optimisation of the light readout system in the ArDM experiment [12].

2. WLS Coating Techniques and Reflectors

2.1. Reflector type

Two materials, ESR (Vikuiti™ Enhanced Specular Reflector foil) from the company 3M and Tetratex™ (TTX) from the company Donaldson Membranes were investigated. 3M foil is a multilayer specular reflecting polymer film and as such is likely to be of high radio-purity. Its appearance is that of a polished metal although the material is non conducting by its nature. It has a specular reflection coefficient of approximately 100% in a large region of the optical spectrum. TTX is an aligned polytetrafluoroethylene (PTFE) fibrous cloth and is nearly a 100% diffuse lambertian reflector.

Because of the method of manufacture, which relies on extrusion of polymer chains within an oil based emulsion, doubts have been raised concerning both the purity of TTX and its outgassing rate. In order to investigate the radio-purity of TTX, samples were sent to Harwell Scientics for analysis by inductively coupled plasma mass spectroscopy (ICP-MS). TTX radiopurity was found to be $1.0 \pm 0.3$, $< 0.4$ and $< 500$ ppb for U, Th and K concentrations respectively. The outgassing properties were checked with a dedicated sample of 48 g of 254 µm TTX (equivalent to 1/20 of the total amount required to line the ArDM experiment). The sample was placed within a large chamber which was evacuated. A series of measurements confirmed the good outgassing properties of TTX.

2.2. TPB deposition techniques on substrates and PMT windows

TPB powder can be applied to a reflector or PMT window by vacuum evaporation, spraying, or by dissolving in a polymer matrix [7, 8]. Vacuum evaporation was performed in an Edwards model E308 evaporation chamber. TPB powder, which has a melting point of 207 °C, was heated electrically in the vacuum chamber by applying 24 A current to a molybdenum sample holder containing up to 3 g of powder. The reflector/PMT window was placed above the TPB powder at a fixed distance and the coating thickness was controlled by varying this distance and the weight of the powder. Sprayed coatings were prepared by dissolving TPB in toluene in a ratio of 1 to 40. This solution was then airbrushed onto the substrate using 1.2 bar argon gas. The polymer matrix coatings were prepared using long chain paraloid or polystyrene plastic fragments dissolved in toluene. An amount of TPB was added and dissolved isotropically. A known amount of liquid was then syringed onto the substrate. The TPB concentration within the solution was varied, as was the amount of liquid applied to the substrate. The solution was left for three hours to allow the toluene to evaporate, forming clear TPB impregnated plastic.

3. Experimental Procedures and Results

3.1. Reflection coefficient measurements of TPB coated reflectors

An Optronic OL750 spectroradiometer with an OL740-70 integrating sphere [13] has been used to measure reflectance and global fluorescence from 200 nm to 700 nm for a range of samples deposited by spray, evaporation, and polymerisation over a variety of thicknesses and concentrations and on a range of substrates. For a full description of the the experimental setup see [12, 14].

The main results of the experiment are shown in Figure 1. The wavelength on the horizontal axis refers to the incident light only, selected by the monochromator. Reflection coefficients greater than one indicate fluorescence. The reflection coefficient for 3M foil decreases approximately by 5% as the TPB coating thickness drops from 4.0 mg/cm² to 0.1 mg/cm² at 450 nm. The maximum reflectance coefficient for TPB coated 3M foil is $0.95 \pm 0.013$ at 430 nm.
and corresponds to 4.0 mg/cm² thickness. Whereas, the reflection coefficient is invariant, within error, with coating thickness for evaporated TTX samples and is 0.97 ± 0.014 at 430 nm.

### 3.2. Global efficiency of wavelength shifting and reflection with distance

A schematic illustration of the argon gas apparatus which was used is shown in Figure 2. This apparatus consisted of a sealed polyvinyl chloride (PVC) tube containing a 3 inch uncoated PMT (electron tube type 9302KB). An α source located at the centre of a TPB coated reflector disk was placed within the tube at a fixed distance from the PMT (the total deposition of alpha energy occurs within 4 cm in 1 bar of gaseous argon). Samples of either 3M foil or TTX cloth coated with TPB were placed around the interior walls of the tube. A delivery tube was inserted into the PVC chamber and argon (1 ppm impurity) gas at 1 bar flowed throughout the apparatus. The argon flow rate was used to control the argon purity. Measurements were taken for varied TPB thicknesses between 0.2 mg/cm² and 4.0 mg/cm², which were deposited both via evaporation and spraying. Additionally, the distance between the α source and the PMT was altered in order to investigate the effect of both the attenuation of light following multiple reflections and the reduction in direct VUV light incident on the PMT. The number of photoelectrons collected at the PMT for each separation (defined as the total area of the light pulse) was then plotted against the slow component decay time ($\tau_2$) for various distances $d$. 

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**Figure 1.** Total reflectance of (left) single thickness TTX cloth; (right) TPB evaporated onto 3M™ foil. The insets show an expanded scale in the wavelength range 415 to 575 nm. Taken from [12].

**Figure 2.** A schematic illustration of the argon gas apparatus used to determine the wavelength shifting and reflection efficiency with distance. Taken from [12].
Figure 3. Total photoelectrons for 3200 ns purity against separation from the α source to PMT for TPB coated reflector walled tube. Figure taken from [14].

The results of the analysis are presented in Figure 3. The reduction in the total light collection with increasing distance was found, within errors, to be independent of TPB thickness and substrate. Evaporated coatings on 3M foil consistently underperformed irrespective of coating thickness compared to TTX cloth. Thicker coatings on 3M foil yielded higher light collection whereas light collection from TPB coated on TTX substrates was found to be almost independent of thickness. The 0.2 mg/cm² TPB on TTX yielded within errors an identical result to the 1.0 mg/cm² coating.

3.3. Wavelength shifting direct light incident on the PMT
Figure 4a shows the argon gas apparatus for direct light measurements. The experiment consisted of a sealed PVC tube containing a 3 inch coated PMT. The PMT window was coated with TPB powder with thicknesses ranging from 0.02 mg/cm² to 2 mg/cm² via evaporation, spraying and application of a polymer matrix containing TPB. The sides and base of the PVC tube were covered with 3M foil reflector coated with 1 mg/cm² TPB powder by evaporation. TPB coated reflector walls were used as the ability of the window coating to shift VUV light is equally important as its ability to allow shifted visible light from the walls to penetrate. An α source was positioned 10 cm away from the PMT window and argon gas was flowed continually. The effect of various PMT window coatings on the total light collection was then recorded by plotting the slow component decay time ($\tau_2$) against the total number of photoelectrons collected at the PMT.

Figure 4b presents the results for the optimum PMT coatings. The optimum thickness was 0.05 mg/cm² by evaporation, improving the total light collection by 38%±3.4% at 1000 ns purity compared to that collected with no PMT coating.

4. Conclusions
TPB can efficiently shift the VUV argon scintillation light into the visible blue high quantum efficiency region of low cost borosilicate windowed bialkali photomultiplier tubes. TTX cloth was found to be a preferable reflector when compared with 3M™ foil due to its better light yield and
large tolerance for the TPB layer thickness. Furthermore, the best TPB deposition method was found to be vacuum evaporation which avoids crystallisation and coating inhomogeneities. Based on spectroradiometer measurements, TPB coated TTX was found to have 97% ±1.4% reflectance at 430 nm for all coating thicknesses. The optimum coating and reflector combination was found to be between 0.2 mg/cm² and 1.0 mg/cm² TPB deposited via evaporation on TTX cloth. The best PMT window coating was found to be 0.05 mg/cm² TPB deposited via evaporation.

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