

3D-printing of polystyrene-based scintillator granulates for particle detectors

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Plastic scintillators are widely used in particle physics experiments. Additive manufacturing techniques allow the production of parts with free shapes and, depending on the application, direct integration with other detector components. This opens up new possibilities for the development of, for example, trigger and veto systems or 3D-segmented detectors like high-granularity calorimeters utilizing structured scintillators with diffuse reflective subdivisions. ARBURG Plastic Freeforming (APF) devices feature the processing of several different granulates at the same time including in-line drying, melting points up to 350°C and high-frequency droplet discharging. The usage of granulates to 3D-print plastic scintillators has the advantage that original materials produced without plasticizers or polymerization starters can be used. However, it must be investigated whether the materials degrade under the high process temperatures to which they are exposed. Achieving high transparency and surface quality are further challenges, as with other techniques. Using the APF process, we have 3D-printed scintillator samples made from granulate based on polystyrene. We have used both commercial granulate with POPOP and p-terphenyl wavelength-shifting additives as well as self-made granulate with PPO and bis-MSB. With these samples we have performed several measurements to evaluate their performance with regard to transparency, fluorescence behavior, decay time and light-yield. We present the results by comparison with reference scintillators and polymethylmethacrylate samples.

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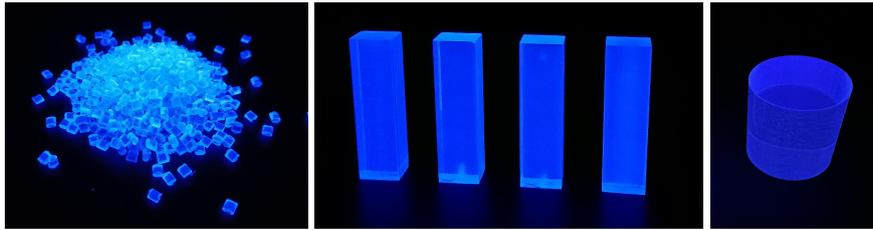


Figure 1: Photograph of the scintillator granulate (left), and four bars (middle) and a cylinder (right) 3D-printed from it. The size of a single granule is in the order of a few millimeters, the length of the bars is 45 mm, and the diameter of the cylinder is 24 mm. Here, all samples are excited with UV light.

1. Introduction

Additive manufacturing ("3D-printing") could become a new way to manufacture plastic scintillators. For example, the ability to print free forms from multiple materials simultaneously is very promising for structured multichannel particle detectors like high-granularity calorimeters [1]. An ARBURG freeformer 300-3X [2, 3] is used for this work, directly processing granulates by melting the material and discharging hundreds of droplets per second through a 200 μm nozzle. This has already been applied by us to three transparent original granulates including polymethylmethacrylate (PMMA) [4]. Extending the process to polystyrene-based scintillators involves that the material is heated up to temperatures above 200°C for more than 10 minutes. Investigating whether this poses a problem for the wavelength shifters and measuring the light yield of the 3D-printed scintillators are the main subjects of the studies presented here. Other 3D-printing techniques, for comparison, require an additional step to produce suitable scintillator filament [5, 6] or resin [7, 8] first.

2. Sample manufacturing

The scintillator granulate purchased is Nuvia SP32 [9]. It is blue emitting (see figure 1, left) and based on polystyrene with p-terphenyl and POPOP additives. We have also produced self-made granulate by mixing STYRON™ 686E with 1 wt% PPO and 0.04 wt% bis-MSB¹. To adjust the printer settings, a qualification is performed for each material. Among others, this procedure is used to reduce air inclusions which decrease the transparency. More details can be found in [4], including transparency measurements for pure STYRON™ 686E. Bar-shaped and cylindrical samples have been printed from Nuvia SP32 granulate and post-processed by milling and polishing (see figure 1, middle and right). As a reference for the measurements described below, samples with the same shape have been purchased².

3. Spectrometer measurements

For the measurement of scintillator emission spectra and decay times the FS5 spectrofluorometer system is used [10]. A xenon arc lamp in combination with a monochromator before and after the sample³, respectively, enables emission scans for different excitation wavelengths. The wavelength

¹Using a rotary mixer and exploiting electrostatic adhesion. Next, the material was extruded and granulated again.

²These references were produced by the manufacturer using cast polymerization of styrene with the same additives.

³One of the bar-shaped samples is inserted into a front-face holder with an inclination to avoid direct reflections.

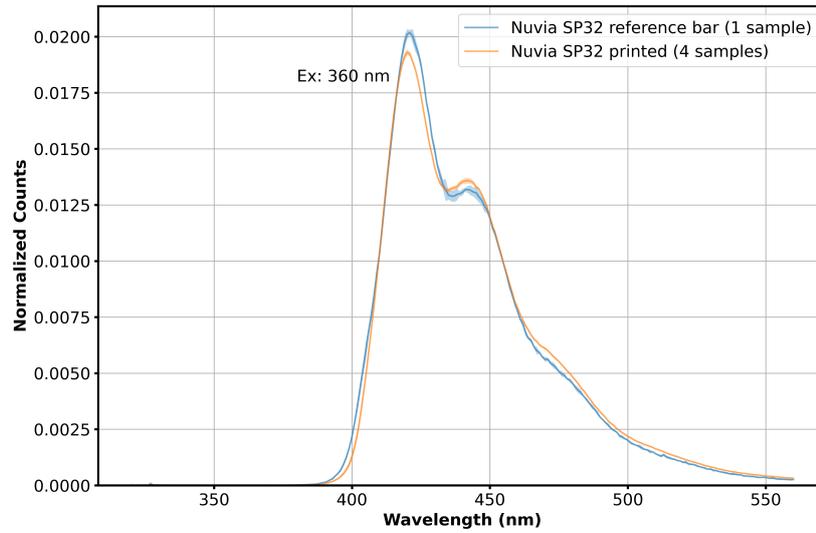


Figure 2: Normalized emission spectra of Nuvia SP32 obtained from spectrometer measurements using an excitation wavelength of 360 nm and 1 nm steps for the emission scans. The blue data shows the spectrum of the reference bar, and the orange data represents the average spectrum of four 3D-printed bars. Each bar is measured at four different surface positions, and the light colored bands represent 95% confidence intervals.

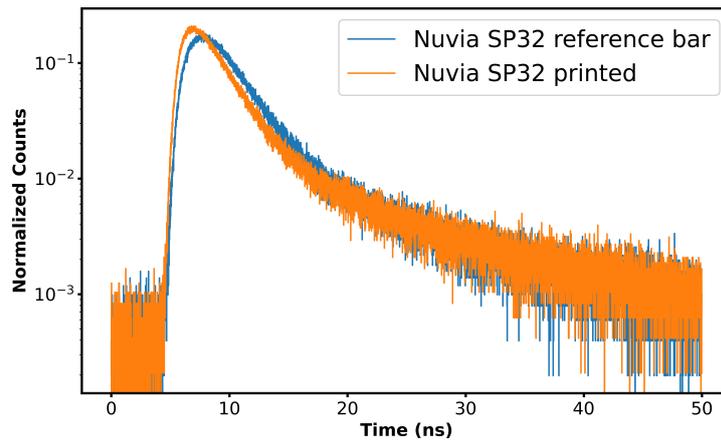


Figure 3: Normalized decay time measurements of Nuvia SP32. A pulsed LED with a wavelength of 255 nm has been used for excitation, with a pulse width of about 900 ps and at a rate of 5 MHz. The blue data shows the results for the reference bar, and the orange data the results for a 3D-printed bar.

coverage is 230-870 nm. Results for Nuvia SP32, presented in figure 2, exhibit the typical POPOP peaks, both for the reference and the 3D-printed samples. Figure 3 shows decay time measurements, using a pulsed light source instead of the xenon lamp and the method of Time-Correlated Single Photon Counting (TCSPC). The monochromator after the sample has been set to select the main POPOP emission peak at 420 nm. A fast and a slow component are visible⁴. Further discussions on the measurements presented here can be found in section 5.

⁴A fit including the instrument response function results in time components of about 3 ns and 16 ns.

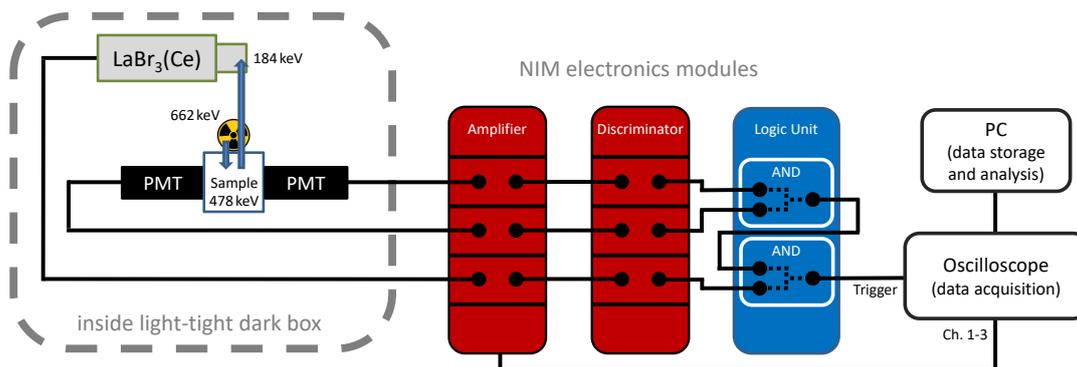


Figure 4: Schematic drawing of the experimental setup to measure the light yield of the samples. The left part is contained inside a light-tight dark box, as indicated by the dashed line.

4. Light yield measurements

In order to measure the light output of the 3D-printed scintillator samples, a dedicated setup is used (see figure 4). The cylindrical sample or reference is placed inside a holder below a ^{137}Cs source, and a $\text{LaBr}_3(\text{Ce})$ detector above. In addition, a PMT is coupled to both end faces of the sample. A triple coincidence trigger is required for events to be stored. In an offline analysis, the Compton 180° backscatter peak is selected in the spectrum of the $\text{LaBr}_3(\text{Ce})$ detector. Thus, in these events mostly 662 keV gammas from the ^{137}Cs source interact in the scintillator sample by Compton Scattering, with 478 keV deposited in the sample and 184 keV deposited in the $\text{LaBr}_3(\text{Ce})$ detector⁵. The signals of the two PMTs, sensing the sample's scintillation light, are integrated and summed up to obtain spectra as shown in figure 5. Determining and comparing the position of the 478 keV peak, a light yield of 39.6% is obtained for the 3D-printed sample relative to the reference⁶. For the reference, a light yield of 56% relative to anthracene is specified, corresponding to ~ 9700 photons/MeV [9]. This has been cross-checked with another scintillator reference (EJ301) to verify the experimental setup.

5. Discussion and conclusions

The results presented in the previous sections show that 3D-printing of polystyrene-based plastic scintillators using APF works. No apparent damage due to the high process temperatures is visible in the measurements taken with Nuvia SP32 granulate. Both the emission spectra and the decay time components of the 3D-printed samples are very similar compared to the reference (see figures 2 and 3). If there was any damage, one would expect that the energy transfer in the scintillator would be disturbed and, for example, the characteristic POPOP peaks in the spectrum

⁵The response of this detector was measured with different radioactive sources to be linear in the range from a few hundred keV to almost 2 MeV.

⁶The light yield uncertainty due to the fit uncertainty of the peak position is $\pm 0.09\%$, the uncertainty due to repositioning the $\text{LaBr}_3(\text{Ce})$ detector and selecting the Compton backscatter peak is $\pm 1.3\%$. Furthermore, a correction is applied to account for the slightly different emission spectra of the sample and the reference (see figure 2) folded with the PMT quantum efficiency. The effect of the correction on the light yield is $+0.3\%$ here.

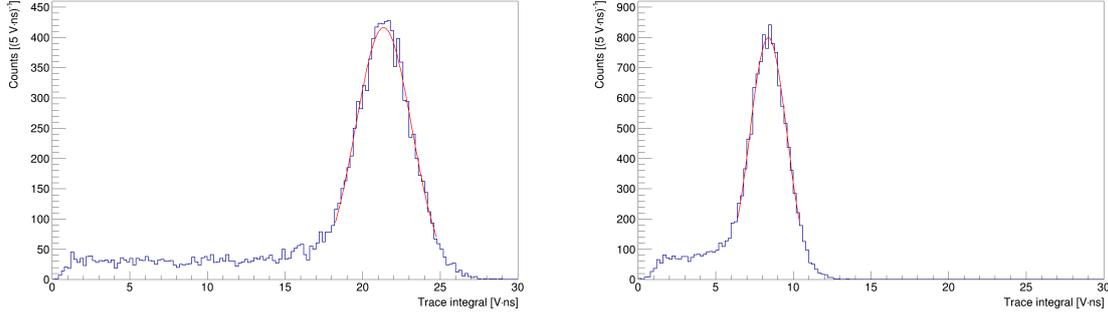


Figure 5: Integrated PMT signals recorded with the light yield setup, both for the Nuvia SP32 reference cylinder (left) and a cylinder 3D-printed from Nuvia SP32 granulate (right). The respective peak corresponds to an energy deposit of 478 keV from a ^{137}Cs source (see text), and the peak position shift between reference and sample is analysed (red fit curves).

would change. The reduced light yield is probably mostly a transparency effect, since our previous studies showed that the transmittance of 3D-printed polystyrene samples is relatively low in the visible and UV range⁷. The spectral shift to higher wavelengths could also be an effect of this. There could also be increased quenching, which would explain the fact that the 3D-printed scintillator samples are slightly faster than the reference. Estimating the amount of quenching is very difficult because, as previously stated, transparency effects probably dominate. Additionally, measurements with first samples 3D-printed from the self-made granulate were made. However, the corresponding emission spectra indicate that the mixing is not completely homogeneous, as besides the bis-MSB peaks also peaks from PPO are visible. Thus, there seem to be local clusters of additives close to the surface. The light yield was measured to be 51.3% compared to the reference⁸.

6. Outlook

Several improvements and new implementations will be tested concerning the process technology. First of all, the transparency of the 3D-printed samples could be increased by optimizing the print parameters and trying different polystyrene granulates. We are currently testing dynamic light scattering to evaluate the usability of granulates for the printing. Furthermore, instead of compressed air, nitrogen could be used for the in-line drying and for flushing the build volume of the printer. In this way, contact of the heated scintillator with oxygen would be reduced. Alternative post-processing methods could be employed and measurements with polarized light could be used to check for stress. In parallel to the process development, we have also started to print application-specific detector parts, in particular structured scintillators which fit in an existing test stand [1]. This will allow for an independent assessment of the light yield as part of a particle detector⁹,

⁷Especially below 400 nm where, for example, PMMA samples 3D-printed with the same APF process reach transmittance values that are at least 20% higher [4].

⁸Here, the light yield uncertainty due to the fit uncertainty of the peak position in the integrated PMT spectrum is $\pm 0.12\%$, the uncertainty due to repositioning the $\text{LaBr}_3(\text{Ce})$ detector and selecting the Compton backscatter peak is $\pm 2.7\%$, and the correction taking into account the emission spectrum is -6.3% . Please remember that the additives for the self-mixed granulate are PPO and bis-MSB, instead of p-terphenyl and POPOP which are contained in Nuvia SP32.

⁹By including both 3D-printed and conventionally produced scintillator tiles, a direct comparison is possible.

enable long-term studies, and demonstrate the flexibility of the printing approach. In particular, the possibilities to implement inlays or structures to guide the light to the photosensors could be very interesting. We will also try to improve the production of self-made granulate, especially the mixing with additives, and to produce self-made references by solvent casting.

Acknowledgments

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