

DEVELOPMENT OF A TEST SETUP FOR A MULTIPIXEL ORGANOMETALLIC IONIZATION DETECTOR

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1. Introduction

The ionization detector is one of the oldest and simplest devices in physics for the detection of ionizing radiation, such as gamma or beta rays. The most common ionization detector consists of a gas-filled chamber in which an electric field is applied, generated by two differently charged electrodes called anode and cathode. An incoming radiation particle now generates electron-ion pairs in the gas, which drift apart due to the electric field and generate a measurable current between anode and cathode. A simple and wellknown example of a detector based on this principle is the Geiger-Müller counter.

In liquid ionization detectors, as the name suggests, a liquid is used as the medium instead of a gas. This has the advantage that the density of liquids is significantly greater than that of gases, which in turn also increases the absorption capacity for the ionizing particles. One difficulty that occurs with liquid ionization detectors, however, is that they have to be ultra-purified in order to be free of any electronegative impurities. For example, the concentration of oxygen in the liquid must be reduced to a level below 1 ppm so that the free path of electrons is large enough to drift from cathode to anode [Gru+08].

A popular choice for the medium of the liquid ionization detector are liquefied noble gases such as argon, xenon or krypton. These have the advantage that, in addition to the ionization signal, scintillation light can also be measured, which is why detectors containing liquefied noble gases are often built in the form of time projection chambers (TPCs). By combining the two signals, a three-dimensional reconstruction of the particle interaction can be generated in the TPC. An example of such a TPC is the XENON100 experiment [Apr+12] and its successors XENON1T and XENONNT, which use liquefied xenon to search for dark matter. A drawback of detectors filled with liquefied noble gases is that they have to be operated at cryogenic temperatures, which usually leads to a more complex and sophisticated design of the detector. However, this disadvantage can be avoided by using materials that are already liquid at room temperature and have electrical properties similar to liquefied noble gases. Suitable candidates are, for example, organometallic liquids such as tetramethylsilane (TMSi). TMSi has often been used in literature as a medium for liquid ionization detectors, for example in the Karlsruhe KAS-CADE experiment, where cosmic-ray hadrons were detected [Eng+99].

The properties of organometallic filled liquid ionization detectors make them also interesting for positron emission tomography (PET) applications. This includes the BOLD-PET project, which aims to build a PET detector based on the organometallic liquid trimethylbismuth (TMBi). TMBi efficiently converts the 511 keV photons in PET into relativistic photo-electrons which in turn create secondary electrons and Cherenkov photons. By detecting the electron signal with a pixelated anode, information about the energy and the interaction position in the material can be obtained.

The objective of this master thesis is to built an ionization detector with a pixelated anode for the use with organometallic liquids and to test whether signals, that one would expect in PET, can be measured. However, before going into detail about the detector, the basics of PET will be discussed in the first section in order to motivate the BOLD-PET project and the construction of the detector.

2. Physical basics of PET

Positron Emission Tomography (PET) is a nuclear imaging technique used in medicine. The technique makes use of the special property of the (low-energy) annihilation of an electron with its antiparticle, the positron (see fig. 1): two oppositely directed photons with the same energy are created. Through simultaneous detection of those two photons, a line can be drawn on which the annihilation must have taken place. These lines ultimately form the database for generating a PET image.



Figure 1: Feynman diagram showing the annihilation of an electron e^- with a positron e^+ , resulting in the creation of two photons γ . Time flows from the bottom to the top.

2.1. Positron emitters and radiotracers

In contrast to other medical imaging methods such as Magenetic Resonance Imaging (MRI) or Computed Tomography (CT), the images generated with PET are not direct depictions of the patient's internal tissue, organs and bones, but rather show the activity of the metabolism in the tissue. For this, however, a positron source must first be brought into the body and the metabolism of the patient. The positron sources used are unstable nuclides that have an increased number of protons compared to neutrons and therefore undergo β^+ -decay. β^+ -decay is a process of the weak interaction in which an up quark (u) of a proton (p) in the nucleus changes its flavour and becomes a down quark (d), which in turn turns the proton into a neutron (n). When the up quark changes its flavour, a W⁺ boson is emitted, which immediately decays into a positron (e⁺) and an electron neutrino (v_e), as can be seen in fig. 2. For an element Y this means that it changes into the isobar X: the mass number A remains the same, while the atomic number Z is reduced by one. The processes on the different atomic levels can be described by the following

reaction equations:



Figure 2: Feynman diagram of the β^+ -decay: an up-quark u inside of a proton p changes its flavour into a down-quark d under emission of a W⁺ boson. Thus the proton becomes a neutron n, while the W⁺ boson immediately decays into an electron neutrino v_e and a positron e⁺.

In eq. (1), however, it should be noted that the decay of the proton does not apply for a free particle and is only possible within an atomic nucleus. The reason for this is the greater mass of the neutron compared to the proton. In order to overcome this difference in mass, the proton must receive energy from outside, which is the case in the nucleus in form of binding energy. The energy released during the β^+ -decay is distributed between the daughter isotope, the positron and the neutrino, resulting in a continuous spectrum from zero to a maximum endpoint energy for the positron.

Table 1 lists some common positron emitters that are frequently used for PET, as well as their half-lives, mean and maximum positron energies and the range of the positrons in water. As can be seen, the half-life $T_{1/2}$ of most positron emitters is relatively short. Since the quality of PET images increases with higher activity and the activity in turn decreases exponentially with time according to

$$A(t) = A_0 \exp\left(-\ln(2)\frac{t}{T_{1/2}}\right),$$
(2)

the short half-lives make it necessary to keep the transport time and distance of the positron emitter to the patient as short as possible. Therefore, the radionuclides are often

Table 1: Properties of common positron emitters used in PET. Values for half-lives and positron energies taken from [Cen]. Values of the (experimental) positron range have been taken from [CLL07], which in turn references to and interpolated the data from [Cho+75]^[1] and [Der+93]^[2].

	Half-life	Mean	Maximum	Mean
Isotopo		positron	positron	positron
Isotope		energy	energy	range in
		[keV]	[keV]	water [mm]
110	20.364 (14) min	385.70 (44)	960.4 (10)	0.92 ^[2] /
C				$1.095^{[1]}$
¹³ N	9.965 (4) min	491.82(12)	1198.5(3)	$1.39^{[1]}$
¹⁵ 0	122.24 (16) s	735.28 (23)	1732.0(5)	$1.785^{[1]}$
18 _E	109.770 (5) min	249.8 (3)	633.5(6)	0.54 ^[2] /
Г				0.9 ^[1]

produced on site in a cyclotron. An example is the production of ¹⁸F: In the cyclotron, a target consisting of ¹⁸O enriched water (H_2 ¹⁸O) is bombarded with protons with an energy of approximately 10 MeV[HCS86]. This leads to a ¹⁸O(p, n)¹⁸F reaction:

$$^{18}\text{O} + \text{p} \rightarrow ^{18}\text{F} + \text{n.}$$
 (3)

The 18 F dissolved in the H₂ 18 O can then be separated chemically (e.g. by distillation) or the solution can be further processed as a whole. How other radionuclides are generated and processed can be found in [WW09]. In order to carry out PET measurements with positron emitters produced in the cyclotron, they first have to be built into a metabolically active molecule, the so-called radiotracer. The radiotracer is usually administered intravenously to the patient and then accumulates at the points in the body that are to be examined by PET, thus ensuring that the positrons are emitted in the regions of interest. There are a number of different tracers that can be used for various processes in the body, most of which are marked with one of the positron emitters from table 1. One of the reasons for this is that the elements carbon, nitrogen and oxygen are the main components of many biomolecules and a stable isotope can easily be exchanged for a positron-emitting one without impairing the function of the molecule. In the case of fluorine, which, in contrast to the other elements mentioned, does not or hardly occur in biomolecules, use is made of the fact that it can often be exchanged for a hydroxyl group (OH) in a molecule without significantly changing its function. This is also the case, for example, with the tracer fluorodeoxyglucose (18 F) (abbreviated [18 F)FDG), which is the most used tracer for PET. [¹⁸F]FDG is structured like glucose (C₆H₁₂O₆), but the hydroxyl group at the C-2 position has been replaced by the positron-emitting ¹⁸F, as can be seen in fig. 3. Despite this small difference, [¹⁸F]FDG in the human body is transported by glucose transporter proteins into cells as if it were normal glucose. Within the cell, the analogy between the two molecules initially continues and the enzyme hexokinase phosphorylates [¹⁸F]FDG and glucose. In the next step, the phosphorylized glucose is broken



Figure 3: Representation of glucose and fluorodeoxyglucose (¹⁸F) in the Haworth projection. The ¹⁸F substitutes a hydroxyl group (OH) at the C-2 position (the bond at the second carbon atom clockwise from the oxygen atom).

down into pyruvate during glycolysis. However, this process requires the hydroxyl group, which in [¹⁸F]FDG is substituted by ¹⁸F. As a result, the phosphorylated [¹⁸F]FDG can no longer be processed in the cell and since it also can not leave the cell in this state, it is metabolically trapped and accumulates there. This process is shown schematically in fig. 4. Thus, with [¹⁸F]FDG, the glucose metabolism in the body can be tracked. Since tumor cells have an increased consumption of glucose compared to healthy cells, these can be made particularly clearly visible with [¹⁸F]FDG, which is why the main area of application for this tracer is oncology.

As soon as the ¹⁸F breaks down to ¹⁸O and a free hydrogen atom is taken up from the environment, the hydroxyl group previously substituted by ¹⁸F is formed, thus the [¹⁸F]FDG becomes normal glucose and can be processed in glycolysis.



Figure 4: Metabolism of glucose (G) and fluorodeoxyglucose (FDG) (redrawn from [Kob+12]). Both molecules are transported into a cell by glucose transporters (GLUT) and, using adenosine triphosphate (ATP), are phosphorylated by the enzyme hexokinase to G-6P (glucose-6-phosphate) and FDG-6P (the ATP becomes adenosine diphosphate (ADP)). G-6P is then further processed into pyruvate in glycolysis, whereas FDG-6P is metabolically trapped in the cell and accumulates.

2.2. Electron-positron-annihilation

As already mentioned, in the β^+ -decay the positrons are ejected from the nucleus with a continuous spectrum of energies which, depending on the positron emitter, can end at several hundred keV to a few MeV. Until annihilation with an electron occurs, the positrons dissipate their energy through inelastic collisions with bound electrons in the surrounding tissue. In-flight annihilation of a positron with an electron is also possible, but the probability of this is very low at approximately 2%[Har04]. The distance that the particles travel until they thermalize and combine with their antiparticles is called positron range. The positron range is a physical limit for the spatial resolution that can be achieved with PET. Since it is dependent on the energy of the positrons, it is therefore also dependent on the radionuclide used and goes up to 1-2mm for the most frequently used positron emitters, as also listed in table 1. One way in which the positron range can be reduced and the spatial resolution improved is demonstrated in [RHC96] and consists in applying a static magnetic field, as is the case with combined MRI-PET scanners. Due to the magnetic field, the Lorentz force acts on all positrons that do not move exactly parallel to the direction of the magnetic field. As a result, the positrons move on helical paths, the radii of which depend on their energy components perpendicular to the magnetic field and on the strength of the magnetic field.

After a positron is thermalized, it can annihilate directly with a valence electron of opposite spin in the surrounding matter, whereby two antiparallel photons are generated, each with an energy that corresponds to the rest mass m_e of the positron or electron respectively $m_e c_0^2 = 511 \text{ keV}$ (c_0 : speed of light). Another possibility is that the two particles form a hydrogen-like bonded state called positronium (Ps). The fraction of positrons that form positronium after thermalization depends on their energy and the material in which they thermalize. For example, a positronium yield of 38% was determined for 4 MeV positrons in water [Har04]. In theory, the formation of positronium can be described by two mechanisms. The first approach is the spur process, in which the positron binds coulombically with an electron that was liberated from the surrounding tissue during thermalization on the path (spur) of the positron. The second approach is the Ore Gap mechanism, in which the positron combines with a bound electron. However, this process may only take place if certain conditions apply to the energies of the particles involved [Yul99]. The energy E_{Ps} of the positronium after formation results due to conservation of energy in

$$E_{\rm Ps} = E_{\rm e^+} - E_{\rm Ion.} + B_{\rm Ps},$$
 (4)

where E_{e^+} is the remainder of kinetic energy that the positron still has when after thermalization, $E_{Ion.}$ is the ionization energy needed to separate the bound electron from a molecule in the surrounding material and $B_{Ps} = 6.8 \text{ eV}$ is the binding energy of positronium. The energy of the positronium is restricted by $E_{Ps} < B_{Ps}$ since a greater energy would result in the quick dissociation of the bond between positron and electron. In regards to eq. (4) this also limits the positron energy to $E_e < E_{Ion.}$. Furthermore there is also a minimum energy for the positron $E_e > (E_{Ion.} - B_{Ps})$ given by the fact that the energy of the positronium must be greater or equal zero. This results in a range of energies for the positron, the so-called Ore-Gap $E_{Ion.} > E_{e^+} > (E_{Ion.} - B_{Ps})$, in which positronium can be formed.

Positrons and electrons belong to the fermions and have a spin $S = \frac{1}{2}$, which means that their z component can assume the values $S_z = \pm \frac{1}{2}$. The positive value corresponds to an upward facing spin state $|\uparrow\rangle$, the negative correspondingly to a downward facing state $|\downarrow\rangle$. When the positron and electron form a bound state, the total spin of the newly formed positronium can either assume the value zero or one. The state with S = 0 is a singlet, since here the z component can only take the value zero, and is called parapositronium (p-Ps). In the S = 1 state, the z component can assume the values 1, 0, and -1, which makes it a triplet and is called ortho-positronium (o-Ps). The possible spin states in which positron and electron can combine to form positronium are thus:

$$|S = 0, S_{z} = 0\rangle = \frac{1}{\sqrt{2}} (|\uparrow\rangle|\downarrow\rangle - |\downarrow\rangle|\uparrow\rangle),$$

$$|S = 1, S_{z} = 1\rangle = |\uparrow\rangle|\uparrow\rangle,$$

$$|S = 1, S_{z} = 0\rangle = \frac{1}{\sqrt{2}} (|\uparrow\rangle|\downarrow\rangle + |\downarrow\rangle|\uparrow\rangle) \text{ and}$$

$$|S = 1, S_{z} = -1\rangle = |\downarrow\rangle|\downarrow\rangle.$$

(5)

Since every spin state is equally likely in the formation of positronium, three times more ortho-positronium than para-positronium is produced.

p-Ps and o-Ps differ in their lifetimes and in the number of photons they decay into due to conservation of charge parity. The charge parity of positronium is $C_{Ps} = (-1)^S$ and thus depends on its spin *S*, i.e. whether it is p-Ps or o-Ps. Since the charge parity of a photon is $C_{\gamma} = -1$, the number *n* of photons into which the positronium decays must follow

$$C_{\rm Ps} = C_{\gamma} \iff (-1)^S = (-1)^n. \tag{6}$$

Para-positronium with $C_{p-Ps} = 1$ therefore decays into an even number of photons and has a vacuum lifetime of 125 ps. The decay into two photons is the most frequent, higher orders are strongly suppressed. The two photon decay corresponds to the case when positron directly annihilates with an electron without formation of positronium, so that due to the conservation of energy and momentum, the two photons are each emitted with an energy of 511 keV and an angle of 180° to each other. Ortho-positronium with $C_{o-Ps} = -1$ must decay into an odd number of photons and has a vacuum lifetime of 142 ns. Because of conservation of momentum the minimum number of photons is three, higher orders are again strongly suppressed. In the case of the o-Ps decay, in contrast to the decay of the p-Ps, the photons no longer have a specific angle to each other and the energy of each photon is no longer the same. Since o-Ps makes up 75% of the positronium formed and 38% of all positrons form positronium (in water), one could assume that the proportion of three-photon-events in PET is at least 28.5%. As a matter of fact, the proportion of decays with more than two photons is only about 0.003% [CD04]. This is because the ortho-positronium interacts with the surrounding matter, which leads to two photon annihilation. On the one hand there can be spin-flip processes, whereby either the spin of the positron or the electron in the o-Ps is reversed and the o-Ps becomes p-Ps with the significantly shorter lifetime. On the other hand,

there can be a pick-off process in which the bound positron reacts with another electron with opposite spin from the environment, resulting in a two photon annihilation. These two processes cause the lifetime of ortho-positronium in matter to be in the range of 0.5 to 5 ns [CSS02], for example 1.8 ns for water [Har04]. It is therefore much more likely that o-Ps in matter will also decay into two photons through interactions with the environment than that it will survive until it decays into three photons like in vacuum.

As mentioned before, the principle of PET is based on the case of two photon decay. If both particles are detected simultaneously in a ring of detectors, a line can be drawn between the two detection points on the ring. Because of the photons are emitted with an angle of 180° between them, one knows that the annihilation must have taken place somewhere along the line. However, this angle is only valid in the rest frame of the positronium, i.e. when the positron and electron have no momentum or kinetic energy. In the laboratory frame, the positronium can have kinetic energy E_{Ps} and a momentum $p_{Ps} = 2\sqrt{E_{Ps}m_e}$ after its formation. This momentum is transferred to the photons when the positronium decays, so that in addition to their momentum of $m_e c_0$ in the rest frame, they each receive a component of $\sqrt{E_{Ps}m_e}$ parallel to the direction of the positronium. As a result the photons are no longer emitted at an angle of 180° to each other, but instead deviate from it by an angle of 2θ , as can be seen in fig. 5. This effect is called noncolinearity. The angle θ is given by

$$\tan\theta = \frac{\sqrt{E_{\rm Ps}m_{\rm e}}}{m_{\rm e}c_0} = \sqrt{\frac{E_{\rm Ps}}{m_{\rm e}c_0^2}}.$$
(7)

The angular distribution with which the photons are emitted is approximately Gaussian in shape with a FWHM of $2\theta \approx 0.5^{\circ}$ [CD04]. By rearranging eq. (7) into

$$E_{\rm Ps} = m_{\rm e} c_0^2 \tan^2 \theta \,, \tag{8}$$

one can get the corresponding kinetic energy of the positronium, which gives $E_{Ps} \approx 9.7 \text{ eV}$.



Figure 5: If the positronium has kinetic energy when it decays, the photons receive a momentum component parallel to the direction of movement of the positronium. As a result, they are no longer emitted at an angle of 180° to one another, but deviate by an angle of 2θ from it. This is known as noncolinearity.

The noncolinearity leads to an error in the localization of the annihilation, since in PET it is always assumed that the photons are emitted back-to-back. This error is given by

$$\Delta = R \tan \theta \tag{9}$$

where *R* is the radius of the detector ring. For a human PET scanner with a radius of approximately 40 cm and θ = 0.25°, this results in an error of 1.75 mm.

2.3. Photon detection

Not all photons emitted are also detected by the PET scanner. In order to be counted by the detectors, the photons have to deposit their energy in the detector material through interactions. However, these interactions only take place with a certain degree of probability, which is described by their cross-section. Two types of interactions are of interest for PET, the photoelectric effect and the Compton effect.

The photoelectric effect is the interaction favored for PET, since the photon deposits all of its energy in the detector. All of its energy is transferred to an orbital electron of an atom. The electron is ejected from the atom and the resulting hole in the electron shell is filled by another electron under emission of characteristic X-rays. The cross-section of the photoelectric effect decreases with increasing photon energies, but shows resonances at energies at which another shell can be ionized. Furthermore, the cross-section increases proportionally with approximately Z^5 [Leo94], which is why materials with a high *Z* are favored for PET detectors.

In the Compton effect, the photon only transfers part of its energy E_{γ} to an electron and is scattered by an angle φ . The energy E'_{γ} of the photon after scattering and the energy

 $E_{\rm e}$ of the electron are dependent on the scattering angle φ and can be described by

$$E'_{\gamma}(\varphi) = \frac{E_{\gamma}}{1 + a(1 - \cos\varphi)} \quad \text{and} \tag{10}$$

$$E_{\mathbf{e}}(\varphi) = E_{\gamma} - E_{\gamma}'(\varphi) = E_{\gamma} \left(1 - \frac{1}{1 + a\left(1 - \cos\varphi\right)} \right),\tag{11}$$

with $a = E_{\gamma}/(m_e c_0^2)$. The maximum energy transfer takes place at a scattering angle of 180° and results for 511 keV photons in an electron energy of 340.67 keV, as can be seen in fig. 6. This maximum electron energy is called the Compton edge, because in the Compton effect, in contrast to the photoelectric effect, one does not measure a sharp energy peak, but a spectrum that is continuous up to the Compton edge. The spectrum can be derived from the Klein–Nishina formula, which defines the differential cross section of photons scattered at an electron [KN29]:

$$\frac{d\sigma}{d\Omega} = \frac{r_{\rm e}^2}{2} \frac{1}{\left(1 + a\left(1 - \cos\varphi\right)\right)^2} \left(1 + \cos^2\varphi + \frac{a^2\left(1 - \cos\varphi\right)^2}{1 + a\left(1 - \cos\varphi\right)}\right).$$
 (12)

with the classical electron radius $r_e = e^2/(4\pi\epsilon_0 m_e c_0^2) \approx 2.82 \times 10^{-15}$ m. As shown in [Joh+52], the cross section can also be expressed as a function of the electron energy:

$$\frac{\mathrm{d}\sigma}{\mathrm{d}E_{\mathrm{e}}} = \frac{\mathrm{d}\sigma}{\mathrm{d}\varphi} \left(\frac{\mathrm{d}E_{\mathrm{e}}}{\mathrm{d}\varphi}\right)^{-1},\tag{13}$$

with

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\varphi} = 2\pi \sin \varphi \frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} \,. \tag{14}$$

The derivative of the electron energy with respect to the scattering angle results from eq. (10):

$$\frac{\mathrm{d}E_{\mathrm{e}}}{\mathrm{d}\varphi} = E_{\gamma} \frac{a\sin\varphi}{\left(1 + a\left(1 - \cos\varphi\right)\right)^2}.$$
(15)

If one now inserts eqs. (12), (14) and (15) into eq. (13), one gets

$$\frac{\mathrm{d}\sigma}{\mathrm{d}E_{\mathrm{e}}} = \frac{\pi r_{\mathrm{e}}^2}{m_{\mathrm{e}} c_0^2 a^2} \left(2 + \frac{s^2}{a^2 \left(1 - s\right)^2} + \frac{s}{1 - s} \left(s - \frac{2}{a}\right) \right),\tag{16}$$

with $s = E_e/E_{\gamma}$. The Compton spectrum of the electrons described by this equation can be seen in fig. 6. The cross-section of the Compton effect is proportional to *Z*.



Figure 6: Top: energy of photon E_{γ} and electron $E_{\rm e}$ as a function of the scattering angle φ in the Compton effect according to eq. (10) for an incident photon energy of 511 keV. Bottom: Compton spectrum as a function of the electron energy according to eq. (16) for a photon energy of 511 keV.

In addition to the cross-section σ , the mass attenuation coefficient μ/ρ is also often used to describe how probable an interaction is. These two are related via $\mu/\rho = N_A \sigma/m_a$, where N_A is Avogadro's number and m_a is the atomic molar mass. Figure 7 shows the mass attenuation coefficient for three liquids: water (H₂O), tetramethylsilane (Si(CH₃)₄) and trimethylbismuth (Bi(CH₃)₃). Water is chosen because the photons can interact not only in the detector, but also in the human body and water makes up a large part of it. The other two liquids are chosen because they are of interest for this work as materials for liquid ionization detectors and will be discussed in more detail in the next chapter.



Figure 7: Mass attenuation coefficient in dependence of the photon energy for water (H_2O) , tetramethylsilane $(Si(CH_3)_4)$ and trimethylbismuth $(Bi(CH_3)_3)$. In addition to the interactions relevant for PET (Compton effect and photoelectric effect), the components of Rayleigh scattering and pair production are also shown. Data taken from [Ber+].

If two photons interact simultaneously in two different modules of a detector ring, it is assumed that they were created in the same annihilation process. A line of response (LOR) which contains the point of annihilation is then drawn between the detector modules. A picture of the activity in the body can later be reconstructed from these LORs. However, the resolution of the reconstructed image is influenced by the fact that incorrect LORs are also recorded, as shown in fig. 8. On the one hand, the photons can scatter in the body by the Compton effect, so that the LOR does not describe the actual path of the photons. These wrong LORs can, however, be filtered out if the detector has a good energy resolution, since the photons lose energy during the Compton scattering. On the other hand, two photons that do not originate from the same annihilation can be detected simultaneously. The number of these random coincidences can be reduced if the detector has a good timing resolution, which allows for a smaller coincidence window. If the timing resolution is in the sub-nanosecond regime, it is even possible to make timeof-flight (TOF) measurements, which improve the image quality further. In TOF measurements, the time difference between the detection of the two photons is measured, by which the point of annihilation can be limited to a certain range along the LOR.



Random coincidence

Figure 8: Different types of coincidences in PET measurements. Tru coincidence: the point of annihilation lies on the LOR. Scattered coincidence: one photon underwent Compton scattering, the annihilation is not located on the LOR. Good energy resolution can filter these events out. Random coincidence: two photons from unrelated annihilations are detected simultaneous. Good timing resolution allows for a smaller coincidence window and thus a smaller number of random coincidences.

Another factor that worsens the resolution of the reconstructed image is the parallax effect, shown in fig. 9. This effect becomes greater the further the annihilation is located from the center of the detector ring. If the photons do not enter the detector modules perpendicularly, it is possible that they pass through a module without interaction but are detected in the adjacent module. This shifts the LOR towards the center of the detector. This can be corrected if the detectors are able to measure the depth of the photon interaction.



Figure 9: Parallax effect: At the edge of the detector ring, a photon can pass through a detector module without interacting, but can be detected in the neighboring module. The resulting LOR is then misplaced towards the center of the ring.

3. Ionization in liquids

As can be seen from the previous chapter, a PET detector should have the following properties:

- a high atomic number to effectively convert photons into photo electrons,
- a good energy resolution to discard scattered events,
- a good timing resolution to discard random events and allow time-of-flight measurements and
- the ability to measure the depth of interaction to improve spatial resolution.

A detector that meets all of these criteria is being developed in the BOLD-PET in the form of a liquid ionization detector.

3.1. BOLD-PET

The BOLD-PET project (Bismuth Organic Liquid Detector) is a collaboration between the scientists from CEA Saclay, France, and the Institute of Nuclear Physics and the European Institute of Molecular Imaging from the University of Münster. It is based on the CaLIPSO detector (fig. 10) (Calorimetre Liquide Ionisation, Position Scintillation Organometallique) proposed by the Saclay group [Yvo+14] and aims to build a liquid ionization detector filled with the organometallic liquid trimethylbismuth (TMBi, (Bi(CH₃)₃)) for PET applications. TMBi is a transparent, insulating liquid with a density of $2.3 \,\mathrm{g/cm^3}$ and contains 82% by weight of bismuth. Thanks to the high atomic number of bismuth (Z=83), a photo fraction of 47% is expected for 511 keV photons. The generated photo-electrons are relativistic in TMBi and thereby generate Cherenkov radiation. Besides that, the photo-electrons generate further electron-ion pairs in the liquid. This allows for two readout channels, an optical one and an electrical one. The optical channel can be read out with photomultipliers which gives a fast timing signal. The electrical channel can be read out by applying an electrical field and drifting the electrons through a Frisch grid to a pixelated anode, from which the energy and the x-y-position of the photon interaction can be reconstructed. From the difference in the timing signal and the electron signal the z-position of the interaction can be reconstructed, therefore enabling a full depth of interaction reconstruction. Overall, through the combination of optical and electrical readout it is anticipated to reach a timing resolution better than 150ps and a depth of interaction resolution in the detector volume of 1 mm³ [Ram+16].

The development of the two readout channels in the BOLD-PET project is divided between the Saclay group and the Münster group. The Saclay group works on the readout of the optical signal, while the Münster group works on the electrical readout. In this context, this work comes into being, in which a multipixel liquid ionization detector is developed. However, for first measurements this detector will not be filled with TMBi, but rather with another organometallic liquid, tetramethylsilane (TMSi). The reason for this is that TMBi must be free of any electronegative impurities in order to obtain a usable signal-to-noise ratio for electrical read out. At the time of this work, the cleaning processes of the TMBi are still in progress, which is why TMSi is used to benchmark the detector. TMSi is in contrast to TMBi a well-known material and has already been used successfully for a liquid ionization detector in the Münster laboratory [Pet19]. Nevertheless, the multipixel detector is designed in such a way that it can also be filled with TMBi later.



Figure 10: Sketch of the CaLIPSO detector (redrawn from [Far+18]). An incident photon creates a relativistic photo-electron in the TMBi which in turn creates Cherenkov radiation and secondary electrons and ions. The Cherenkoy photons are detected with PMTs while the electrons drift towards a pixelated anode due to an electric field and induce a signal when they pass a Frisch grid.

3.2. Onsager theory

In liquid ionization detectors, electron-ion pairs are generated by high-energy particles; in the case of PET, these are photons. The electrons then transfer their energy through in-

elastic collisions to neighboring molecules until they thermalize. If they move far enough away from their parent ion during this process so that the Coulomb attraction is less than the thermal energy of the liquid, the electrons can diffuse into the volume and recombine with another ion. If this is not the case, the electron can recombine again with its parent ion, also known as initial recombination. The critical distance r_c , at which the Coulomb energy is equal to the thermal energy, is given by

$$r_c = \frac{e^2}{4\pi\epsilon_0\epsilon_r k_B T},\tag{17}$$

with ϵ_r the relative permittivity of the liquid and *T* the temperature. Onsager considered the initial recombination in the presence of an external electric field *E* and created a theory that describes the escape probability of the electron in this case [Ons38]. Since the derivation of this theory is relatively complex, reference is hereby made to [Sch97], in which this is dealt with in detail. For low electric field strengths, the escape probability is given by

$$P(r, E, T) = \exp(-r_c/r) \left(1 + \frac{e^3}{8\pi\epsilon_0\epsilon_r k_B^2 T^2} E \right) = \exp(r_c/r) \left(1 + \frac{e}{2k_B T} r_c E \right).$$
(18)

From this equation it follows that even without an electric field there is a probability

$$P(r, 0, T) = \exp(-r_c/r)$$
 (19)

that an electron will escape its parent ion. A measure of on many free electron-ion pairs are generated in a liquid by a source is the free ion yield G_{fi} . It is defined as the number of electron-pairs escaping recombination per 100 eV of deposited energy. Analogous to the Onsager theory, the dependency of the free ion yield on the electric field strength can be described by

$$G_{fi}(E) = G_{fi}^0 \left(1 + \frac{e^3}{8\pi\epsilon_0\epsilon_r k_B^2 T^2} E \right), \tag{20}$$

with G_{fi}^0 the free ion yield at zero field strength. The zero field free ion yield G_{fi}^0 for TMSi has already been measured quite frequently in the literature, also in the Münster laboratory, which resulted in 0.54 (3) [Pet19] and agreed well with other results. As far as is known, the zero field free ion yield for TMBi has so far only been measured by the Saclay group, which resulted in a value of 0.083 (5) [Far+18].

4. Setup of the multipixel oragnometallic ionization detector

The test setup developed for this work consists of the actual multipixel detector and a purification bench with which the detector is filled with the TMSi and which cleans the TMSi. The construction of both parts is described in the following.

4.1. Detector cell

4.1.1. Anode

The detector presented in this thesis consists of a pixelated anode. Free charge carriers that are generated by an external radioactive source are drifted through the chamber and detected with a charge-sensitive preamplifier. A 58 mm × 58 mm *Photonis* MCP-PMT (MicroChannel Plate-PhotoMultiplier Tube) [Pho14; And+08] feedthrough is used as forms of a pixelated anode. It consists of a 1.5 mm thick ceramic plate with 32 × 32 metal pads that are feed-through the plate. The two sides of the plate differ in the size of the pixels as can be seen in fig. 11. The side facing inwards in the MCP-PMT has a pixel size of 1.4 mm with a pitch of 1.6 mm, as well as a 2 mm wide metal layer on the edge of the anode. The outward-facing side on which the readout electronics are attached has a smaller pad size of 1.1 mm with the same pitch and no metal edge.



(a) Inwards facing side of the multipixel an- (b) Outwards facing side of the multipixel ode anode

Figure 11: The two sides of the ceramic multipixel anode. The inwards facing side has a pixel size of 1.4 mm with a pitch of 1.6 mm and a 2 mm wide metal ring, the outwards facing side has smaller pixels of 1.1 mm with the same pitch.

In addition to forming the electrical field necessary to drift free electrons, the multipixel anode also forms an integral part of the detector housing. Pressing the anode with a fixating brace seen in fig. 12a against a flange and using a gasket to seal the connection a vacuum-tight housing can be achieved (for the read out of the anode a printed circuit board is put between anode and fixating brace, which is discussed in more detail in section 5). Due to the size of the anode, the flange is a modified standard CF flange DN 63 made of stainless steel, which has a frame for the anode and the fixating brace with M5 threaded holes on one side. The fixating brace is also made of stainless steel and has five holes (16 in total) along each side, from where it can be attached to the flange with M5 screws. As of the writing of this thesis the multipixel anode presented is the only prototype in existence. To reduce the risk of damaging this prototype on assembly, test measurements using ceramics of similar dimensions are conducted. These measurements yield that the screws used to adjust the pressure exerted by the fixating brace to the flange can be tightened with up to $12 \text{ N} \cdot \text{m}$ without breaking, when following the order depicted in fig. 12c.



Figure 12: (a) Using M5 screws a fixiating brace is pressed against the detector board and multipixel anode to compress a gasket that seals the anode against the flange. The Anode is an integral part of vacuum tight housing of the detector. (b) Ceramic plate that is used to test at which torque the multipixel anode could break when tightening the screws of the fixating brace. (c) To reduce the mechanical stresses exerted on the ceramic plate and later the multipixel anode, the screws of the fixating brace are tightened in a pattern shown here. By always tightening the screws opposed to each other results in an even pressure on the ceramic.

Regarding the gasket, a material softer than the anode and the must be used in order to get a good seal and not damage the anode in the sealing process. The detector should

be suitable for TMBi. Therefore, materials such as Viton or Indium, which decompose TMBi, can not be used as a gasket material. For this reason, the decision was made to use gold as the sealing material, which is sufficiently soft and does not react with TMBi. A tight sealing requires the gasket to run on a planar surface. Since the anode has the metal ring on the inwards facing side, there are two transitions in height on this side that can lead to leaks if the gasket crosses them: the transition from metal ring to ceramic and from ceramic to metal pads. The gasket should therefore run either only over the metal ring or over the ceramic, in this case the metal ring is chosen. In order to obtain a gold gasket in the required length and shape, a commercially available gold wire ring with an inner diameter of 69.9 mm and a wire diameter of 0.63 mm is taken and bent into shape. So that the gold wire stays in position, a groove with a width of 0.7 mm and a depth of 0.35 mm is embedded in the flange, which can be seen in fig. 13b.

4.1.2. Cathode

The electrical field in the detector is generated by an electro-polished, stainless steel cathode opposite the anode, to which a high voltage with negative polarity is applied. The high voltage is fed into the detector via an SHV feedthrough, which is welded into a CF blind flange DN 63. The entire detector is grounded via the shield of the feedthrough. Two electro-polished, stainless steel filler blocks are attached to the flange in order to keep the volume of liquid in the detector as low as possible. As a result, the volume of liquid in the filled detector is approximately 30 mL.



Figure 13: (a) Electro-polished filler blocks and cathode attached to CF63 flange with SHV feedthrough. (b) Gold wire gasket in the groove of the frame for the fixating brace.

The cathode is square with a side length of 43 mm, with the corners rounded with a radius of 5 mm, and has a thickness of 3 mm. Attached directly to the pin of the feedthrough, the distance between cathode and the anode is about 4 mm, the active volume between

cathode and anode is thus approximately 7.3 mL. All other distances in the detector to the cathode are kept larger so that the electrical field in the detector is the greatest between anode and cathode. This was checked with a COMSOL simulation, the result of which can be seen in fig. 14a. Since the cathode is smaller than the pixelated area of the anode, the electrical field for pixels at the edge of the anode is not homogeneous, which must be taken into account when reading them out. This can be seen again in a COMSOL simulation of the electric field lines starting from the cathode fig. 14b.



⁽b) Electric field lines

Figure 14: COMSOL simulations of the electric field strength between anode and cathode and the electric field lines starting at the cathode when $-1 \, \text{kV}$ is applied to the cathode and everything else in the detector is grounded.

In order to fill the detector cell with liquid, one of the CF63 flanges is equipped with two *Swagelok* VCR adapters. For first tests, however, only one adapter is required so that the second one is closed with a plug. The second adapter is needed if the detector is to be operated under a continuous flow of liquid.

Figure 15 gives an overview of all components and the structure of the detector cell. Technical drawings of all the components can be found in the appendix (appendix A).



Figure 15: CAD-drawings of all the components of the detector cell (made by Christian Huhmann).

4.2. Purification bench

Filling the detector with TMSi is done by connecting it to a purification bench designed for this purpose, which can be seen in fig. 16. All parts in the bench are made of stainless steel or glass and are connected via Swagelok VCR connections or CF flange connections and thus satisfy ultra high vacuum requirements. The TMSi used was previously used for the measurements by Simon Peters in the Münster laboratory [Pet19], for which it was cleaned through various molecular sieves and then stored in a sealed container under vacuum. Nevertheless, it is cleaned again in this setup with a 4Å molecular sieve. Molecular sieves consist of porous materials with a uniform pore size that filter gases or liquids through adsorption. When molecules that are smaller than the pore size enter a molecular sieve, they penetrate the pores and are trapped there, while larger molecules cannot and leave the molecular sieve again. A 4Å molecular sieve has a pore size of 0.4 nm (= 4 Å)and adsorbs molecules such as water, carbon dioxide, oxygen and small hydro carbons. In order to activate the molecular sieve, it is baked out for three days at 400 °C, whereby all residues of bound molecules (mostly water) in the sieve are released and pumped out. Meanwhile, the rest of the bench up to the glass-to-metal connection is baked out at a temperature of 200°C degrees and pumped via a turbo molecular pump to remove residual gases absorbed on the walls, ending at a pressure in the order of 1×10^{-7} mbar.



Figure 16: CAD-drawing of the purification bench (made by Christian Huhmann).

When the filling process of the detector is started, all valves except the ones for the vacuum pump and the bypass are opened. The bypass is only there to quickly empty the detector in the case of an emergency. The copper clamp below the molecular sieve is connected to a thick copper braid that is immersed in liquid nitrogen, which cools the bench at this point and creates. Under atmosphere, TMSi already evaporates at 26 °C, in vacuum lower temperatures are sufficient. The different temperatures between TMSi reservoir and copper clamp result in different vapor pressures of the TMSi. This creates a pressure gradient through which the liquid evaporates in the warm spot and condenses in the cold spot from where it flows into the detector. This process can be accelerated by slightly heating the TMSi reservoir. Over the whole time, the pressure in the system is monitored with a *WIKA WUC-10* pressure gauge, which can measure overpressure and has a reading accuracy of 0.01 mbar. This is important because TMSi has a vapor pressure of almost one bar at room temperature, which increases sharply when heated. If the pressure becomes too high, the filling process must be interrupted as not all components in the system are suitable for overpressure.

Since the detector itself does not have a viewport, the filling level can be checked via the glass-metal connection. When this is reached, the valve to the TMSi reservoir is closed to end the filling process. The glass-metal connection is mounted below a bellow which reduces mechanical stress on the glass-to-metal transition.

5. Electrical connection of the multipixel anode

The detector is designed in such a way that the pads of the multipixel anode are easily accesable for contacting and the anode can easily be removed again by unscrewing the fixating brace. This was done because the detector should ultimately be read out with the ASIC (Application-Specific Integrated Circuit) IDeF-X HD, which was developed by the Saclay-group. IDeF-X (standing for Imaging Detector Front-end) is a 32-channel analog front-end with self-triggering capability, including a charge sensitive amplifier and peak detector in every channel [Gev+21]. It is planned to use a total of eight IDeF-X, which are each attached in pairs on printed circuit boards (PCBs). As can be seen in fig. 17, these four ASIC boards are connected via cables to a mother board, which mainly functions as a power supply and shares analog and digital signals for parallel communication. The mother board is directly connected to the so-called MATIS board, from which the individual IDeF-X are controlled and read out. Due to the design of the ASIC boards, the connection to the detector must take place via a mini edge card connector (MEC5-050-01-L-DV-W1-K-TR by Samtec). This means that another board is required, which is attached directly to the detector or rather on the multipixel anode and which contacts the individual pads with the connectors.



Figure 17: Schematic drawing of the proposed readout of the the detector using the IDeF-X ASICs. Four ASIC boards, each with two IDeF-X chips, are plugged directly into a detector board via suitable connectors, which is attached to the detector's multipixel anode. The ASIC boards are connected to a mother board and a MATIS board via cables, from where they are supplied with energy and controlled.

Since 256 channels are available with the total of eight IDeF-X, but the anode has 1024 pixels, two possible variants of the detector board were considered: read out only a quarter of the available pixels and area, or use the entire area and combine four pixels into one channel and thus possibly have higher noise¹. Ultimately, the second variant was chosen since it allows to read out a larger area and therefore active volume. The finished board can be seen in figs. 18a and 18b.

¹Combining four pixels increases the area and thus the capacitance *C* per channel, which leads to a higher noise charge $Q_n = \sqrt{k_{\rm B}TC}$.



(a) Side of detector board facing the multipixel anode

(b) Read out side of detector board



(c) Pixelated PCB used to test different connection methods.

Figure 18: Top: detector board which combines four pixels lying on a square into one channel. 32 pixels are lost due to the connectors. For the first tests only three of the four connectors are attached. Bottom: PCB resembling the multipixel anode used to test different connection methods. Both PCBs have a side length of 59 mm × 59 mm, a thickness of 1.65 mm, a pad size of 1.1 mm × 1.1 mm and a pitch of 1.6 mm. Detector board designed by Roland Berendes.

Like the multipixel anode, the detector board has on one side 32×32 pixels with a pad size of $1.1 \text{ mm} \times 1.1 \text{ mm}$ and a pitch of 1.6 mm, which each combine on the other side in groups of four at one channel of the connectors. Due to the connectors, there are eight two by two pixel large blind spots. So that eight channels are not lost in the read out, the pixels at these spots are grouped together in such a way that only three pixels form a channel, so there are 32 channels that consist of only three pixels. With a side length of $59 \text{ mm} \times 59 \text{ mm}$ and a thickness of 1.65 mm, the detector board is only 1 mm wider than the anode and can therefore be easily attached to the detector between anode and fixating brace.

In order to electrically connect the detector board to the multipixel anode, so that all pixels have good contact and there are no short circuits between adjacent pixels, various options were tried out. The common method in industry to create such a connection
would be reflow soldering. In this process, solder paste is applied to the individual pixels before the components are positioned on top of each other and heated in a reflow oven, creating a permanent solder joint. When heating, a special thermal profile is used. Due to different thermal properties of the ceramic and the metal in the anode this can cause mechanical stress on the component and possibly damage it. Therefore reflow soldering and all other connection methods involving heat are not suitable for the anode. In order not to expose the anode to any other dangers during the tests of different connection methods, these are first carried out with test PCBs. The test PCBs, which are shown in fig. 18c, are constructed like the anode and have 1024 through-going pads of size $1.1 \text{ mm} \times 1.1 \text{ mm}$ and pitch 1.6 mm, their size corresponds to the size of the detector board. Unlike the multipixel anode the test PCBs have holes of 0.4 mm at the center of each pad.

5.1. Tests of different connection methods

5.1.1. Anisotropic conductive paste

The first method considered is an anisotropic conductive adhesive (ACA). Like normal conductive adhesives, ACAs consist of an adhesive base material such as acryl or epoxy that is filled with conductive particles. The difference is that in ACAs the proportion of conductive particles is significantly lower, making the adhesive as a whole non-conductive [Opd16]. A conductive connection is only created when the components to be connected are pressed together with the adhesive in between, since the conductive particles are clamped between the pads of the components, as can be seen in fig. 19. Due to their small proportion the material, the particles (almost) do not touch each other, but only the opposing pads in the assembly (vertical axis), which means that there are no conductive connections between neighboring pads (horizontal axes), hence the name anisotropic.



non-conductive direction

Figure 19: Working principle of an anisotropic conductive adhesive (ACA). The ACA (brown) is filled with conductive particles (blue) which are trapped between the opposing pads of the two components (here detector board (green/yellow) and multipixel (black/grey) anode) and thus create an electrical connection between them. However, the proportion of particles in the material is so small that they do not touch each other so there are no conductive connections between adjacent pads. Drawing not to scale.

ACAs are available in the form of prefabricated films (ACFs) and pastes (ACPs) that have to be mixed prior to usage. The advantage of ACFs is that they have a defined thickness and the distribution of conductive particles is uniform. With ACPs, the self-mixing and application can result in areas that have a different thickness or concentration of particles. The disadvantage or limitation of ACFs is that they require high pressure and temperature to bond the two components together, typical values are > 20 bar and > 150°. ACPs, on the other hand, are available with a variety of curing conditions; for example, some can be cured at room temperature or under UV light.

Since high temperatures have to be avoided due to the sensitive ceramic-metal transitions of the anode, the choice fell on the Anisotropic Conductive Epoxy Adhesive 124-19 by Creative Materials, a two part, room temperature curing ACP [Inc16]. The conductivity of this adhesive is created by silver particles. When cured, it has a specified volume resistivity of $0.001 \Omega \cdot cm$ in the vertical axis and $1 \times 10^{12} \Omega \cdot cm$ in the horizontal axes. The curing time is 24h during which a pressure of over 100 psi must be applied, which corresponds to a weight of approximately 200kg based on the pixelated area of the test PCBs. A mechanical press is used to apply this pressure. Since the ram surfaces of the press are smaller than the test PCBs, steel blocks are placed between the rams and the boards to be glued, so that the pressure is transferred to the entire area of the boards. To check the pressure, a load cell calibrated with weights is put in on one side between the steel block and the ram. A sketch of this setup is shown in fig. 20. The load cell outputs a voltage proportional to the pressure or weight, which ensures that this is above the required value over the total curing time. On both test PCBs, the holes in the pads were soldered shut before the ACP was applied, as otherwise the paste would be squeezed out of them when pressed and spread on the other side. The ACP is mixed in the specified weight-ratio of 100:22 between epoxy and curing agent and applied as evenly as possible in a thin layer to one of the PCBs². Then the other board is positioned over it and both are pressed together by hand, whereby excess paste oozes out to the sides and can be removed. Afterwards, the PBCs are put into the press.

 $^{^2} Both$ PCBs were cleaned with isopropanol before the ACP was applied.



Figure 20: Sketch of the setup used to connect two test PCBs with anisotropic conductive paste. A pressure equivalent to over 200kg to the pixaletd area of the PCBs is applied with a mechanical press over a period of 24h at room temperature. Since the press surfaces are smaller than the PCBs, steel blocks are put in between to distribute the pressure evenly. The pressure is checked with a load cell, which is put in between a steel block and the press.

After the ACP has hardened, the electrical contact between the opposing PCBs is checked and whether there are short circuits between adjacent pixels. To do this, the resistance between the connected pads is measured with a digital multimeter (Gossen Metrawatt METRAHIT One). A total of 500 of the 1024 pixels are measured, ten by ten in the corners of the PCBs and ten by ten in the middle. The result of the measurement can be seen in fig. 21. No shorts were found between neighboring pixels, which shows that the ACP is working as intended. Overall, the resistance is on average $0.30(3)\Omega$, although there are differences between the measured locations on the PCBs. The resistance at the pixels in the upper left corner (referring to the position in fig. 21) is comparatively uniform with an average of $0.1354(18) \Omega$. In the lower left corner it looks similar with a mean resistance of 0.148 (4) Ω , but here there are two pixels where the test PCBs have no contact. On the right side there are larger fluctuations between the individual pixels, which leads to a mean resistance of $0.32(3)\Omega$ and $0.39(5)\Omega$ respectively in the upper and lower corners. A total of three pixels measured on the right side are without contact. In the middle there are the largest deviations between pixels, resulting in a mean resistance of $0.59(13)\Omega$, and with a number of eleven also the most dead pixels.



Figure 21: The measured resistance between the pads of two test PCBs bonded with anisotropic conductive paste. A total of 500 of the 1024 pixels are measured, ten by ten in the corners of the PCBs and ten by ten in the middle, the mean values of the various positions are listed above/below them. The systematic uncertainty results from the accuracy of the multimeter used.

The deviations of the measured resistance in the various areas of the test PCBs as well as the presence of pixels without contact suggest that the ACP was not mixed and applied evenly enough, as a result of which the number of conductive particles trapped between the opposing pads varies from pixel to pixel, and/or the pressure required in the curing process was not evenly distributed over the boards. These are factors that can be attempted to improve through further tests with the test PCBs, but there is no guarantee that dead pixels and areas with varying levels of contact will not occur again when connecting the detector board and the multipixel anode. In addition, the connection created with the ACP is permanent and cannot be released again without damaging the two components, which means that the multipixel anode would no longer be usable if the contact was not sufficient. Therefore, in the end, it was decided against ACP as the method for the electrical connection of the detector board and the multipixel anode.

5.1.2. Solder balls

The next consideration for establishing a (reversible) electronic contact between the detector board and the multipixel anode is to use solder balls which are a common connection method used in electronics in so-called ball grid arrays (BGAs). In BGAs, the balls are attached to the underside of a component, which can then be placed on a circuit board and heated in an oven, causing the balls to melt and connect to the pads of the circuit board, creating electrical contact between component and board. Since heating is not an option because of the anode, the idea is to establish electrically contact through the pressure exerted by the fixating brace on detector board and anode. The solder balls are placed on the pads of the anode and the board, aligned accordingly, is pressed from above, so that the balls are clamped between opposing pads. To ensure that there is only one ball on each pad and that it cannot escape to the sides, a 3D printed grid is placed on the anode as a spacer, the height of which is smaller than the diameter of the balls. A schematic drawing of this can be seen in fig. 22.



Figure 22: Schematic drawing of the electrical connection with solder balls (not to scale). The balls (blue) are placed on the pads of the anode and held in place by a 3D printed spacer (red). The detector board is aligned and placed on top of the anode, by applying pressure the balls are clamped between opposite pads and ensure an electrical connection.

This procedure is first tested again with two test PCBs instead of detector board and anode. Two different solder balls with diameters of 0.55 mm and 0.76 mm are tested. The 0.55 mm balls do not need a spacer because they are already held in place by the holes in the test PCBs, as seen in fig. 23. For the 0.76 mm balls, a grid with a hole size of 0.8 mm, pitch 1.6 mm and height 0.4 mm is printed from PLA. For both ball sizes, it is observed that there is contact between the pixels at the edge of the PCBs, where the fixating brace presses onto the boards, but not in the middle. This suggests that the pressure of the fixating brace is not evenly distributed over the surface of the boards, so that the balls that are in the immediate vicinity of the brace are pressed together by the boards, but in the middle the balls are not in contact with the upper board. One explanation for this would be that the solder balls are too hard compared to the boards, so that the PCBs tend to bend rather then flatten the balls under the pressure. This could create a small bulge in the middle of the PCB so that the balls would no longer be clamped tightly between the pads there.





5.1.3. Conductive silicone

Since the solder balls are not suitable as a means of establishing electrical contact due to their relative hardness compared to the test PCBs, another, more flexible material is considered to replace the balls. With more flexibility, the pressure differences and the resulting bending of the board can be compensated by adapting the thickness of the conductive material between the boards to the pressure: in places with higher pressure the material is compressed more than in places with lower pressure. One such material is electrically conductive silicone. However, conductive silicone is not commercially available in the form of small balls that could just replace the solder balls in the setup, but as a paste. To form small balls on each pad from this paste, it is applied by stencil printing to one of the pads. The silicone paste used is CHO-BOND 1075 by Parker Chomerics which achieves a volume resistance of $0.01 \Omega \cdot cm$ due to silver-plated aluminum particles in the silicone and requires a minimum thickness of 0.25 mm and a curing time of one week at room temperature. A 0.3 mm thick copper sheet was used as stencil, in which 32×32 holes with a diameter of 1 mm and a pitch of 1.6 mm were drilled so that the holes are above the pads of the PCB when the stencil is placed over it. The paste is first applied to a point on the copper sheets without holes and then wiped over the holes with a squeegee and pressed through them. The copper sheet is then removed, leaving small pieces of silicone on the pads, that correspond in shape and size to the holes in the sheet. After curing, the silicone is firmly attached to the pads so that the board can be pressed against a second one (shown schematically in fig. 24) and the contact between the pixels can be measured.



Figure 24: Schematic drawing of the electrical connection with conductive silicone (not to scale). The silicone (blue) is applied via stencil printing to the pads of the detector board, which is then aligned and placed on top of the anode. By applying pressure the silicone connects the pads of board and anode, pressure differences over the area are compensated by deformation of the silicone.

In principle, it is found that the method with silicone works better than the solder balls because there is also contact between pixels that are further away from the brace, but there are still areas in the middle of the PCBs without contact. The problem with the silicone, which causes this method to be discarded, is the texture of the paste, which does not allow it to be applied evenly to the pads of the board via stencil printing. It is very difficult to spread the paste over the copper sheet and press it through the holes, where most of it gets stuck and therefore does not settle on the pads of the board. However, if a silicone paste is used that is suitable for stencil printing, this method could lead to good contact between the detector board and the anode.

5.1.4. Conductive silver glue

The stencil printing process used for the conductive silicone can also be used to apply (isotropically) conductive adhesive to the individual pads and thus establish electrical contact, as depicted in fig. 25. The problem is that most of these adhesives, like the ACP, are based on epoxy and are therefore permanent, or that, like the silicone, they are not suitable for the process of stencil printing. Therefore, based on a patent for ethanol-soluble electrically conductive adhesive [Heg15], which is suitable for stencil printing, it is produced in-house. This glue consists of shellac, ethanol and silver powder (see fig. 26). The shellac used is form of flakes and has been dewaxed (otherwise it would not be soluble in ethanol), the ethanol has a purity of at least 99% and the silver powder used has a particle size of 3 to 5μ m and a purity of 99.99%.



Figure 25: Schematic drawing of the electrical connection with conductive glue (not to scale). The glue (blue) is applied to pads of the multipixel anode by stencil printing, the detector board is then aligned and placed on top. After curing, the pads of anode and board are electrically connected.



Figure 26: The three components of an ethanol-soluble electrically conductive adhesive: dewaxed shellac in the form of flakes, ethanol with a purity of at least 99% and silver powder with a particle size of 3 to 5µm and a purity of 99.99%.

In order to achieve the right texture of the adhesive for stencil printing and to achieve good conductivity, several tests are carried out with different mixing ratios of the three components. Ultimately, the best result is achieved with a mixing ratio of 1:1:4 between shellac, ethanol and silver powder. First, shellac and ethanol are mixed together while stirring, whereby heating to approximately $60 \,^{\circ}$ C accelerates this process. When the shellac flakes are completely dissolved, the silver powder is added and mixed in well. After that, the glue can be applied to the board. The copper sheet which was used for the silicone is not used for this, but a stencil made of stainless steel with a thickness of $120 \,\mu$ m that fits the detector board, as can be seen in fig. 27. If the stencil printing is successful and all pads are covered with an adhesive layer, the second test PCB is aligned appropriately and put on top. For the adhesive to harden, it was found that it is sufficient if the PCBs are placed on a smooth, flat surface and are evenly pressed on with a weight of



approximately 10 kg for a period of three days at room temperature.

Figure 27: The stencil used to apply the conductive glue to the boards. It is made of stainless steel and has a thickness of 120µm, its pattern match the design of the detector board (fig. 18a).

As with the ACP, the resistance between 500 pixels of two test PCBs is measured in areas of ten by ten pixels in each corner and in the middle with a multimeter to check the electrical contact that is achieved in this way. No short circuits between neighboring pixels can be detected, which shows that stencil printing is a suitable method to connect detector board and anode. Overall, the measured resistance between the pixels is on average 0.37 (3) Ω therefore very similar to the average resistance of 0.30 (3) Ω of the ACP. As with the ACP, there are also fluctuations between the measured locations on the PCBs with this adhesive. This time the center is most uniform with a resistance of 0.156 (2) Ω . In the corners there are spots of pixels with higher resistance than in the surrounding area, resulting in average resistances of 0.34 (2) Ω , 0.36 (7) Ω , 0.45 (5) Ω and 0.54 (9) Ω (starting from the upper left corner clockwise). In each corner there is also an area of two by two pixels that have no contact. This is because no glue could be applied there due to the stencil, since these areas correspond to the pixels that would be lost through the connectors on the detector board. Otherwise no further pixels can be found without contact.



Figure 28: The measured resistance between the pads of two test PCBs bonded with an ethanol-soluble electrically conductive adhesive. A total of 500 of the 1024 pixels are measured, ten by ten in the corners of the PCBs and ten by ten in the middle, the mean values of the various positions are listed above/below them. The two by two spots of dead pixels are the pixels to which no glue could be applied due to the stencil used. The systematic uncertainty results from the accuracy of the multimeter used.

The places at which the resistance of the pixels is increased compared to adjacent pixels could be explained by the fact that either less adhesive was applied there during stencil printing, or that, due to unevenness, the pressure there deviated slightly from the other places during curing. In contrast to the ACP, however, no places could be found that have no contact as a result of these factors. For this reason, and because the connection can be released again without damaging the boards, it was decided to use this method to connect the detector board and the anode. It should be noted, however, that if the adhesive is to be used for applications that go beyond mere testing, it should first be subjected to qualitative testing. For this purpose, the influences of various factors on the conductivity and adhesive strength of the glue should be tested more closely, for example

the curing time and temperature, the layer thickness or the pressure during curing.

6. Pulse measurement with TMSi

With the choice of the ethanol-soluble adhesive, all preparations are now complete to assemble the detector and to carry out the first test measurements with TMSi.

6.1. Assembly of the detector

in order to carry out first test measurements, the multipixel anode is now connected to the detector board by the ethanol-soluble silver adhesive in the manner described in section 5.1.4, in order to carry out first test measurements. Initially it is attempted to read out only one channel, i.e. four neighboring pixels. For this, a BNC cable is soldered to a channel of the detector board, which belongs to four pixels in the middle of the board, as can be seen in fig. 29.



Figure 29: Detector board with a BNC cable attached to one channel for read out.

The glued together anode and board are placed on the gold wire seal on the detector flange and carefully pressed on with the fixating brace. The screws are tightened very carefully in the order described in section 4.1.1. The screws are initially tightened with a torque of $0.2 \text{ N} \cdot \text{m}$ which is then increased in steps of $0.2 \text{ N} \cdot \text{m}$ up to $1 \text{ N} \cdot \text{m}$. From $1 \text{ N} \cdot \text{m}$ on, the steps are increased to $0.5 \text{ N} \cdot \text{m}$ until a torque of $3.5 \text{ N} \cdot \text{m}$ is reached³. A leak test is carried out after each step. Despite the increase in torque, the measured leak rate stagnates at $1 \times 10^{-3} \text{ mbar} \cdot \text{L/s}$ (a value $< 1 \times 10^{-6} \text{ mbar} \cdot \text{L/s}$ is desired for a leak-tight setup). Therefore it is decided to loosen the fixating brace again and examine whether the gasket

³The increasing step-size is due to a change in tools.

is possibly damaged. It is found that, despite the previous tests with the ceramic plate, all four corners of the anode are broken.

The reason for breaking of the corners can be assumed to be that the contact pressure of the fixating brace was not sufficiently even distributed over the anode. One factor here could be the bond between anode and detector board. While the adhesive was hardening, the two components were pressed together by placing them on a smooth surface and putting a weight of approximately 10kg on the connectors. However, since only three out of four connectors are attached to the detector board, the pressure has not been evenly distributed over the entire surface of the board, so that the adhesive layer between the two components has a non-uniform thickness. This can be seen in fig. 30 and influences how the contact pressure of the fixating brace is distributed over the anode.



Figure 30: (a) Conductive adhesive applied to the pads of the anode by stencil printing before it is bonded with the detector board. (b) Layer of adhesive after disassembling the broken anode and the board. Because the pressure was not applied uniform, the layer thickness varies between the pads.

Another factor that could have contributed to the broken edges of the anode is the pressure transfer at the corners of the fixating brace. In contrast to the detector board, the corners of the anode are chamfered, so the board protrudes slightly at these points. When the fixating brace presses on the detector board and the anode, the pressure there is distributed differently between these two parts, as seen in fig. 31 on the left. This can lead to a kind of leverage effect, in which the part of the board protruding over the anode acts as a lever arm and the gold wire acts as a fulcrum. This additionally increases the force acting on the anode at these points, which could ultimately cause the breakage. The broken corners of the anode are reattached with a vacuum-compatible epoxy adhesive (*Thorlabs TS10*) so that measurements can still be carried out. The adhesive creates small differences in height that lead to leaks when sealing, the glued-on corners are polished flat on the inward-facing side of the anode (seen in fig. 32). Furthermore, the break points run over the metal ring of the anode, which means that it can no longer be guaranteed that the gasket underneath will be leak-tight. The gasket is therefore moved further inwards on the detector flange so that it no longer goes over the metal ring but rather the ceramic part of the anode. To ensure even pressure distribution the next time the fixating brace is tightened, the detector board is removed and the screws at the corners of the brace are omitted. Without using the detector board, a BNC cable is soldered to a single pixel of the anode for read out. A narrow frame made of PE is placed between fixating brace and anode to ensure that the pressure is applied precisely to the area of the anode above the gasket, to avoid leverage effect, as shown in fig. 31 on the right.



Figure 31: Left: schematic sketch of the pressure distribution (orange arrows) of the fixating brace at the corners of the assembly. Since the corners of the anode are chamfered, the detector board protrudes slightly so that the pressure is not transferred evenly over the anode. Right: pressure distribution at the new assembly. Since the PE frame between anode and fixating brace is directly above the indium wire, the pressure is evenly transferred to it.



Figure 32: Anode repaired with vacuum-compatible epoxy adhesive. Because the adhesive creates small height differences, the inwards facing side (left picture) is polished flat. For the new gasket further inside, an indium wire is used instead of a gold wire, as indium is softer and might give a better seal. However, this means that the detector in this form is no longer suitable for TMBi, since TMBi can react with indium but also with the adhesive used to fix the anode. The measurements presented in the following are therefore meant as a proof-of-concept of the read out of charge pulses on individual pixels of the multipixel detector. In contrast to the gold wire gasket, the indium wire gasket does not consist of a closed ring, but rather has two ends that flow into one another during the sealing process. There are several ways to join the ends of the wire to create a good seal, as shown in fig. 33. Since there is only little space available for the gasket and the wire has a diameter of 1 mm, the method of cutting both ends to a point and placing them very close to each other is chosen. This can be seen schematically in the middle panel of fig. 33 and in the left picture of fig. 34a in practice.



Figure 33: Different ways to join the ends of an indium wire to create a good seal. The ends can be overlapped, which makes the gasket initially thicker at this point, which can lead to a different pressure distribution when it is first tightened. If the difference in height is not desired, the ends can be cut sharply and placed in front of one another so that they flow into each other when tightened. In this case, however, it can happen that the ends are too far apart and the gasket is not tight as a result. To prevent this, the wire can be laid twice, but this requires more space.

The detector is reassembled with the new indium gasket in the same procedure as with the gold gasket, seen in fig. 34b. The screws are tightened up to a torque of $2.5 \text{ N} \cdot \text{m}$, at which a leak rate of 1×10^{-4} mbar·L/s is measured. Tests with a leak checker suggest that the detector side containing the joint of the two ends of the indium wire is responsible for the observed leak. Since this leak rate is still so high that there is a risk of oxygen entering the detector from the outside and contaminating the TMSi, an argon filled bag is placed around the detector.



Figure 34: (a) New indium wire gasket further inside. (b) Assembled detector with a BNC cable soldered to one pixel for read out.

6.2. Electronics

In PET measurements, a detector must be able to measure signals caused by individual 511 keV photons. In order to test this for the detector created in this work, an energy spectrum is measured in the presence of a radioactive source and compared with a spectrum without source. The radioactive source used is ¹³⁷Cs with an activity of approximately 1.2MBq which produces mono-energetic photons of 661.7 keV and is placed directly on the flange of the detector during measurement, as seen in fig. 35. The detector is located in a Faraday cage and wrapped in aluminum foil to prevent external electromagnetic fields from interfering with the measurement signal.



Figure 35: Setup for the measurement with ¹³⁷Cs source. The argon-filled bag prevents oxygen from entering detector and contaminating the TMSi.

A drawing of the electronic setup is shown in fig. 36. The cathode of the detector is supplied with a voltage of -1 kV by an *iseg NHQ224M* high voltage power supply during the measurements. The anode, or rather one pixel of the anode, is connected to a charge-sensitive preamplifier. The design of the preamplifier is based on the extremely low-noise preamplifiers used in the former Mainz neutrino mass experiment and was made in-house by the electronics workshop [Sch90]. It amplifies the relatively low charge signal generated in the detector and produces a voltage proportional to it. In addition to the input for the detector, the preamplifier also has an input for a test signal. Here, an *Ortec Model 419* precision pulse generator is connected. It delivers a test pulse with an amplitude of approximately 2.44 mV and a frequency of 50 Hz, with which the dead time correction and signal broadening of the detector can be determined later. The output signal from the preamplifier is fed to a *CAEN Mod N968* spectroscopy amplifier and amplified again with an amplification factor of 200 and a shaping time of 3µs. The signals are then digitized with an *Ortec 926 ADCAM* MCB (**M**ulti**C**hannel **B**uffer) and read in with a PC.





6.3. Results

A spectrum with and without a source is recorded for a duration of 60 min each. For the number of counts *N* measured in each channel, an error of \sqrt{N} is assumed, ccording to the Poisson distribution⁴. In both spectra a noise contribution at low channel numbers (<500), i.e. at low energies, which decreases exponentially is visible, as well as the signals generated by the pulser in the form of a Gaussian peak at higher channel numbers (>1500). To be able to compare the two spectra, a dead time correction must first be carried out. For this purpose, the signal generated by the pulser is used. Since the pulser emits a signal with a frequency of 50 Hz, a total of 50 Hz · 60 min = 180000 counts caused by the pulser should be measured over the entire measurement period. The dead-time correction factor then results from the ratio of expected signals by the pulser to those actually measured. After adding up the measured pulser counts, a factor of 1.027 (2) is obtained for the background measurement and 1.083 (3) for the measurement with source, with which the respective spectra are multiplied. The result is shown in fig. 37.

When comparing the two spectra, one can see that at low channel numbers, the count rate is higher when measuring with source and deviates from the exponentially decreasing noise signal at a channel number of approximately 400. This is what is to be expected, since free electrons are generated in the TMSi by photons emitted by the source, which induce a charge on the anode, and it shows qualitatively that pulses can be measured with the detector. Another difference that is noticeable is that the pulser peak is wider when measuring with the source, but has a slightly lower amplitude. This could be due to an increased dark in current in the detector induced by the source and caused by the ions generated alongside the free electrons in the TMSi.

⁴At small count numbers (N < 10), the \sqrt{N} error is no longer valid. In order to simplify data processing, however, it is still assumed here.



Figure 37: Measured data without (upper panel) and with source (middle panel) for a measuring time of 60 min. With the help of the pulser peak, a dead time correction can be carried out. After this correction and normalization to the measurement time, the count rates of both measurements can be directly compared (bottom panel).

In order to also check quantitatively whether the deviation of the two spectra can be ascribed to charge pulses created by photons from the ¹³⁷Cs source, it must first be discussed what kind of spectrum is expected. As already mentioned, the background spectrum $N_{\text{background}}(x)$ can be described by an exponentially decreasing noise distribution and the Gaussian pulse peak:

$$N_{\text{background}}(x) = A_{\text{exp}} \exp\left(-\frac{x}{t}\right) + A_{\text{gauss}} \exp\left(-\frac{(x-\mu)^2}{2\sigma^2}\right),$$
(21)

where x is the channel number, t the decay constant of the exponential part, μ and σ the position and width of the Gaussian part and A_{exp} and A_{gauss} are the respective amplitudes of both parts. Fitting this equation to the background spectrum, as seen in fig. 38, gives the values $A_{exp} = 1.14 (4) \times 10^6$ cps, t = 18.31 (5), $A_{gauss} = 282.4 (8) \times 10^{-3}$ cps, $\mu = 2059.80 (17)$ and $\sigma = 70.36 (13)$ with a reduced chi-squared χ^2 /ndof = 1239.18/777 = 1.59 (5). The relatively large deviation from the ideal reduced chi-squared of 1 can mainly be explained by two factors: On the one hand, the electrical noise deviates from the exponential function for small channels, which is why a further component would have to be added to the fit for a more precise description. On the other hand, the pulser peak is not exactly described by a Gaussian, because as is visible from the data, the two edges are not exactly symmetrical.



Figure 38: Measured background spectrum with a fit according to eq. (21).

In order to describe the measurement with the source, a term must be added to eq. (21) that describes the signal induced by the source. For an ideal detector with unlimited energy resolution where the energy of the incident photons is completely transferred to an electron due to the photo-effect, this term would be a delta peak at the energy E_0 of the source, which can be seen in fig. 39 in the upper left picture. However, a real detector has a limited energy resolution that can be described by a Gaussian uncertainty, visible in the upper right picture in fig. 39 and mainly responsible for the observed width of the pulser peak. Another effect that occurs with liquid ionization detectors is the dependence of the signal on the location of the photon interaction in the detector. As Simon Peters showed in his master thesis [Pet19], the charge *Q* induced by the generated free electrons depends on the interaction position *z* between anode and cathode:

$$Q = -e\left(1 - \frac{z}{d}\right),\tag{22}$$

where *d* is the distance between anode and cathode and *z* can range from 0 to *d*. Since it must be assumed that the photons interact everywhere between cathode and anode with the same probability, this leads to a constant spectrum between 0 and E_0 for a monoenergetic source, as seen in lower left picture of fig. 39. This position dependency can be eliminated with a Frisch grid. The grid is attached directly in front of the anode with a slightly lower potential so that electrons only induce a charge on the anode when they pass the grid.

For the liquid ionization detector, the expected signal for photo-electrons from a monoenergetic source is now given by the convolution of the Gaussian uncertainty with the position dependence, which can be seen in the lower right picture of fig. 39. The convolution of two functions f(t) and g(t) is defined as

$$(f * g)(t) = \int_{-\infty}^{\infty} f(\tau)g(t-\tau)d\tau.$$
(23)

If one assumes a step function of the form

$$f(t) = \begin{cases} 1 & \text{if } t \le E_0 \\ 0 & \text{otherwise} \end{cases}$$
(24)

for the position dependence, and for the Gaussian uncertainty

$$g(t) = \frac{1}{\sqrt{2\pi\sigma}} e^{-\frac{E_0^2}{2\sigma^2}},$$
 (25)

this gives for the convolution of the two

$$(f * g)(t) = \frac{1}{\sqrt{2\pi\sigma}} \int_{-\infty}^{E_0} e^{-\frac{(E_0 - \tau)^2}{2\sigma^2}} d\tau.$$
 (26)

By substituting $z = (\tau - E_0)/(\sqrt{2}\sigma)$ and $dz = d\tau/(\sqrt{2}\sigma)$, eq. (26) can be written as

$$= \frac{1}{\sqrt{\pi}} \int_{-\infty}^{E_0} e^{-z^2} dz = \frac{1}{\sqrt{\pi}} \left(\int_{-\infty}^0 e^{-z^2} dz + \int_0^{E_0} e^{-z^2} dz \right).$$
(27)

The first integral of the sum in eq. (27) is given by $\sqrt{\pi}/2$, and the second one can be expressed using the error function

$$\operatorname{erf}(x) = \frac{2}{\sqrt{\pi}} \int_{0}^{x} e^{-\tau^{2}} \mathrm{d}\tau$$
, (28)

which ultimately gives

$$= \frac{1}{\sqrt{\pi}} \left(\frac{\sqrt{\pi}}{2} + \frac{\sqrt{\pi}}{2} \operatorname{erf}(E_0) \right) = \frac{1}{2} \left(1 + \operatorname{erf}(E_0) \right) \,. \tag{29}$$

The expected signal can thus be described by the error function $\operatorname{erf}(x)$. The counting rate N(x) for the detection of mono-energetic photo-electrons of energy E_0 with a Gaussian uncertainty σ of the detector as shown in fig. 39 can thus be described by

$$N(x) = \frac{N_0}{2} \left(1 - \operatorname{erf}\left(\frac{x - E_0}{\sqrt{2}\sigma}\right) \right)$$
(30)



where N_0 is the amplitude in cps and x is the energy.

Figure 39: Expected signal for a mono-energetic photo-electrons. Top left: measured signal with an ideal detector. Top right: broadened signal with Gaussian energy smearing of a real detector. Bottom left: position dependent signal in a liquid ionization detector. Bottom right: convolution of Gaussian smearing and position dependency.

For the TMSi-filled detector used in this work, a mono-energetic ¹³⁷Cs source with $E_0 = 661.7$ keV is used, but most of the time not the entire energy of the photons is detected. This is due to the fact that with TMSi the proportion of Compton scattered photons is significantly higher than those participating in the photoeffect, which means that even with an ideal detector, not an energy peak, but a continuous spectrum up to the Compton edge at 477.34 keV is measured. For the expected spectrum, this means that the spectra according to eq. (30) must be added up for each individual energy E_i and amplitude N_i in the binned Compton spectrum:

$$N(x) = \sum_{i=1}^{n} \frac{N_i}{2} \left(1 - \operatorname{erf}\left(\frac{x - E_i}{\sqrt{2}\sigma}\right) \right).$$
(31)

The energies E_i and amplitudes N_i of the Compton spectrum can be obtained by simulating the energy deposited in the detector. Since there is no simulation for the detector used in this work, the GATE simulation created by Björn Gerke for Simon Peters TMSi-filled liquid ionization detector [Pet19] is used, which can be seen in fig. 40. However, it

should be kept in mind that due to the different geometry slight changes in the spectra shape between the two detectors occur.



Figure 40: GATE simulation by Björn Gerke of the deposited energy induced by a ¹³⁷Cs source in the TMSi filled detector of Simon Peters [Pet19]. Plotted with 100 bins.

The entire spectrum to be expected for the measurement with source is finally obtained from the sum of eq. (21) and eq. (31), with corrections being applied to eq. (31) in order to take into account that the geometry of the detector used in the simulation deviates from the detector used here:

$$N_{\text{source}}(x) = A_{\text{exp}} \exp\left(-\frac{x}{t}\right) + A_{\text{gauss}} \exp\left(-\frac{(x-\mu)^2}{2\sigma^2}\right) + A_{\text{geom}} \sum_{i=1}^n \frac{N_i}{2} \left(1 - \operatorname{erf}\left(\frac{k(x-b) - E_i}{\sqrt{2}k\sigma}\right)\right).$$
(32)

The correction parameters used are a channel offset *b*, a scaling factor A_{geom} for the count rates and a factor *k* which is applied to the channel number *x* and the Gaussian width σ to convert them into energies since the data obtained by the simulation is in units of energy. Fitting eq. (32) to the measured data with source, as seen in fig. 41, yields $A_{\text{exp}} = 57.8(21) \times 10^3 \text{ cps}, t = 27.82(11), A_{\text{gauss}} = 203.5(6) \times 10^{-3} \text{ cps}, \mu = 2046.15(24), \sigma = 97.34(17), A_{\text{geom}} = 25.4(24) \times 10^{-6}, b = 17(17) \text{ and } k = 1.37(6) \text{ keV}$ with a reduced

chi-squared χ^2 /ndof = 1773.59/1237 = 1.43 (4). The deviation of the reduced chi-squared from the ideal value of 1 can again be explained by the deviation of the noise from the exponential function and the asymmetry of the pulser peak.



Figure 41: Measured spectrum with source with a fit according to eq. (32).

It can be seen that the data is well described by the fit, but the signal expected by the source lies largely in the noise.

6.4. Discussion

The comparison of the recorded spectra with and without the presence of the source shows a clear increase in the counting rate caused by the source, which can also be described by the theoretically expected spectrum. However, since the theoretical spectrum lies largely in the noise, an attempt should be made to reduce the noise further. One way to reduce the noise would be to ground all pixels that are not read out. In the current setup, only one pixel is connected and read out, while all other pixels are on a floating potential, which could contribute to the noise. Nevertheless, it can be concluded that pulses can be seen with the detector when a single pixel is read out. The next step would be to try to read out several pixels at the same time and to see whether a spatial resolution can be achieved by positioning the source at different points around the detector and comparing the spectra of the individual pixels with one another. Regarding the planned readout with the detector board where four pixels are combined to one channel, it is also important to check whether the noise increases too much to be able to measure pulses if multiple pixels are combined to one channel. Furthermore, a simulation of the deposited energy that is adapted to the geometry of this detector should be created in order to be able to compare it with the measured signal and to be able to carry out a channel calibration.

When thinking about an actual TMBi-filled PET detector, two things will greatly help to suppress the observed noise contribution to the spectrum: Firstly, with the large atomic number of bismuth the photoeffect becomes much more probable, so instead of a broad Compton spectrum one can work with a peak of mono-energetic photo-electrons. If the detector contains a Frisch grid, the dependency of the signal on the interaction position is also eliminated. Secondly, as in a PET setup one always measures two photons from an annihilation process, making use of the time coincidence of the two detections one is able to reject the largest part of the background events.

7. Conclusion and outlook

In this work a multipixel organometallic ionization detector was built on the basis of a pixelated ceramic anode with 32 × 32 metal pads. For the read out of the anode, a printed circuit board was designed which is to be attached directly on the anode. For this purpose, various connection methods were tested with PCBs, including anisotropic conductive paste and ethanol-soluble conductive silver adhesive which is applied by stencil printing. With the ACP, an average contact of $0.30(3)\Omega$ could be measured between the pads of two connected PCBs. However, since there were also pads without contact and the connection with ACP is permanent, this method was discarded. For the ethanolsoluble conductive adhesive, a mean contact of 0.37 (3) Ω was measured. Since the connection created with it can be released again and all pads were in contact, this method was used to contact the anode with the board. If the adhesive is to be used for other applications in the future, it is advisable to test it more extensively beforehand and to measure the influence of various parameters on the achieved bonding strength and conductivity. Unfortunately, when assembling the detector, the corners of the anode broke off, which is due to an uneven pressure distribution in the structure. For the next detector, which contains pressure-sensitive components, the pressure distribution should be simulated or calculated beforehand as a precaution. The anode could be repaired and the detector reassembled with slight changes. Since the assembled detector had a leak rate of around 1×10^{-4} mbar L/s, it was put in a bag filled with argon so that there is no danger of oxygen entering the system. Since the leak may be caused by the indium gasket used, the gasket should be checked before the next use of the detector and replaced if necessary. If there is still a leak, another option would be to redesign the detector so that there is a chamber around the anode that is leak-tight and can be pumped out or flooded with argon, so that the temporary bag is not needed anymore.

With the assembled and TMSi-filled detector, an energy spectrum was finally recorded in the presence of a ¹³⁷Cs source for a single pixel read out. By comparing it with a spectrum measured without source one can disentangle the signal and noise contributions. The signal distribution could also be described theoretically, but a large part of it is covered by noise. Therefore it should be tried to reduce the noise in order to get an even clearer signal. Regardless of whether the noise can be reduced or not, the next step is to read out several pixels at the same time in order to obtain a spatial resolution, which is what this detector is ultimately intended for.

A. Technical drawings

All technical drawings shown in this section are made by Christian Huhmann.



Figure 42: Technical drawing of the modified CF63 flange for the multipixel anode.



Figure 43: Technical drawing of the fixating brace.



Figure 44: Technical drawing of the CF63 flange with the SHV feedthrough.


Figure 45: Technical drawing of the filler blocks.



Figure 46: Technical drawing of the cathode.

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