We use positron annihilation lifetime spectroscopy (PALS) in order to study the annealing behavior of aluminum and lead. For the measurement, we use $^{22}$Na as positron source encased with Kapton foil and we utilize a fast-slow delayed coincidence apparatus to record the positron lifetimes. The instrumental resolution curve as determined with a $^{60}$Co source is well-described by a Gaussian with a FWHM of 284 $ps$. We apply this apparatus setup to the positron lifetime experiment of metal samples. We choose aluminum and lead as our samples. We study the annealing behavior of pure aluminum and shop-grade aluminum sample. We found that the annealing temperature for both Al samples lies just below 600$^\circ$C. For pure lead the optimal annealing temperature appears to be near 280$^\circ$C, but the results are not conclusive.
STUDY OF ANNEALING BEHAVIOR OF AL AND PB USING POSITRON ANNIHILATION LIFETIME SPECTROSCOPY

A Thesis

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Faculty of Miami University

in partial fulfillment of

the requirements for the degree of

Master of Science

by

Md Salah Uddin

Miami University

Oxford, Ohio

2016

Advisor: Herbert Jaeger, PhD

Reader: Khalid F. Eid, PhD

Reader: Stephen G. Alexander, PhD

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This Thesis titled

STUDY OF ANNEALING BEHAVIOR OF AL AND PB USING POSITRON ANNIHILATION LIFETIME SPECTROSCOPY

by

Md Salah Uddin

has been approved for publication by

The College of Arts and Science

and

Department of Physics

____________________________________________________
Herbert Jaeger

____________________________________________________
Khalid F. Eid

____________________________________________________
Stephen G. Alexander
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Dedication

To my amazing family, especially to my mom, without whom I would not exist.
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1 Introduction

1.1 Positron annihilation spectroscopy and application

Positron annihilation lifetime spectroscopy (PALS) is a well established, unique, and versatile method to study the nature, concentration and spatial distribution of defects in materials\(^1\text{-}^5\). It is the most sensitive spectroscopic technique among other positron methods, like Doppler broadening, and angular correlation of annihilation radiation\(^6\). PALS is non-destructive to materials due to the small mass of \(e^+\) and the low \(e^+\) current\(^7\). Positrons are used as a probe for the investigation of defects; unlike electrons, positrons are repelled by atomic nuclei and are especially sensitive to vacancy-like defects in solids because of the lack of positive charge in such defects. Thus positrons prefer these sites that have a lower than average electron density. The diffusing positrons are therefore attracted by open volume defects and annihilates preferentially there. As the de Broglie wavelength of positrons at room temperatures is about 2\(\text{nm}\), the thermalized positron “sees” a big portion of the solid – the sensitivity for vacancy-like defects is on the order of 1 \(\text{ppm}\) \(^7,\,^8\). Because of the high sensitivity, PALS can be applied to those cases where the defects and voids are smaller than 10\(\text{nm}\) diameter and PALS is chosen as one of the efficient methods for probing since conventional electron microscopy is inadequate\(^9\).

In a solid, positrons thermalize in about \(10^{-12}\,\text{sec}\) and annihilate in about \(10^{-10}\,\text{sec}\), yielding two or more gamma rays which carry off the momentum of the electron-positron pair. For some materials, a positron-electron bound state called positronium (Ps) is formed. The electron and positron spins may combine to a total spin 0, so-called para-positronium (p-Ps) in a singlet state ((\(^1\)\(S_0\))\(^*\)); alternatively, the electron and positron spins combine to a total spin 1 and the so-called ortho-positronium (o-Ps) in a triplet state ((\(^3\)\(S_1\))); the triplet state (o-Ps) forms three times more likely than the singlet state (p-Ps). The lifetime of a singlet state is 125\(\text{ps}\) and upon decay emits two gamma rays, whereas the lifetime of a triplet state is 142\(\text{ns}\) and decays by emitting three gamma rays each of which has broad distribution of energy ranges from 0 to 511keV. However, the triplet Ps is usually “picked off” by anti-parallel spin electrons, turning the o-Ps into p-Ps, which then annihilates with two gamma rays emission within 125\(\text{ps}\). Different positron states are extracted from a timing spectrum and that can be correlated to structural defects in metals\(^10\), ceramics\(^11\) and semiconductors\(^12\), as well as to nanovoids forming the free volume in polymers\(^13\). Due to those wide variety of applications, PALS is mostly used for the investigation and characterization of solid-state materials\(^14\). Furthermore, the advantage of PALS is that the presence of phase transitions can be conveniently observed by monitoring the positron parameters (lifetime and intensity) as a function of the temperature\(^15\). In all these cases a total number of annihilation events registered in each spectrum of the order of \(10^6\) can be sufficient to obtain the required results. Conversely, higher counts must be accumulated to get a distribution of lifetimes with good accuracy, such as ortho-positronium (o-Ps) annihilation in the nanocavities of a macromolecule\(^16\).

PALS has been used in extremely broad area of materials physics and other areas in science and engineering, like semiconductor physics\(^7\), surface physics\(^17\), polymer physics\(^18\), biophysics\(^19\), positron-atom scattering in gases\(^20\), positron and positronium chemistry\(^21\), and others.

\(^*\)The notation is \(^{2S+1}\)\(L_S\), where \(L\) is the orbital angular momentum quantum number (\(L=0\) is written as S, \(L=1\) as P, etc.), \(S\) is the total spin angular quantum number. The multiplicity of the state is \(2S+1\).
1.2 Motivation of the research

Our intense excitement on positron annihilation lifetime spectroscopy (PALS) lies on its exclusive, detailed information and non-destructiveness to the material. This technique is so powerful that it can be applied to a variety of specific states in solids, ranging from an extended Bloch state in a defect-free lattice to a highly localized state in a vacancy with high sensitivity as the sensitivity of positrons are located at equilibrium vacancy concentration which is $10^{-6} - 10^{-4}$ atomic concentration\[22\]. Because of this unique characteristics of the positron upon annihilation, PALS can yield unique and detailed information regarding with various properties of a material such as information about local electron density and the electron momenta at the site of the defect-trapped positron. Because of the unparalleled behavior of positrons, it can be called a non-invasive probe for understanding the nature of materials. PALS has become a valuable tool for the study of the electronic and defect structures of solids that its application and the physical basis of positron annihilation in condensed matter have been extensively reviewed over time\[23-26\]. So these exercises are the primary basis of our encouragement persuaded us to study the behavior of the positron from the analysis of the lifetime spectrum.

The selection of the sample was a tricky part of the project. Basically post-transition metals are in great interest for scientists because of their various applications. Post-transition metals are the metallic elements in the periodic table located between the transition metals (to their left) and the metalloids. Typically, their crystalline structures tend to show covalent or directional bonding effects and they have generally greater complexity or fewer nearest neighbors than other metallic elements. However, most familiar and popular type of post-transition metals are aluminum, tin and lead (details on aluminum and lead in chapter 2). Besides that, aluminum and lead have lower (13) and higher (82) atomic numbers, respectively, in the periodic table. The lifetime spectrum depends largely on atomic number, $(Z)$ that is shown in Fig 1.2.1 where $I_{\text{Kapton}}$ is the relative intensity of the positrons are passing through the source supporting Kapton foil \[27\]. Therefore, understanding the material with the lower and the higher atomic number will help us to get a sense of the characteristics of the rest of the atomic species.

Primarily we choose two state trapping model which was developed by Brandt and Stewart\[28\], Bergersen and Stott\[29\] and Connors and West\[30\], so it is sometimes called the Brandt-Bergersen-Stott-Connors-West trapping model. It simply states that when the vacancy concentration, which is an exponentially increasing function of temperature (e.g., for aluminium\[31\]), exceeds a certain value essentially every positron is trapped at a vacancy before annihilation\[31\]. However, the theory does not provide information about the redistribution of the electron in the vacancy, but implies about the local lowering the concentration of electron, hence the positron lifetime increases. Mechanical deformation can cause increase in positron lifetime and this characteristic has been tentatively attributed to similar trapping effects at dislocation cores\[32-33\]. Therefore, vacancy and dislocation effect have greater interest in material characteristics\[34\]. The two state trapping model nicely work for aluminum and there are numerous research groups applied this model for their understanding\[35-38\]. Therefore, it is a great impact that the model can be used for the sample in our system and because of abundant literature support we would be able to understand more vividly about our system.

![Figure 1.2.1](image-url)
Primarily we think of annealing and quenching the sample. Annealing is a very popular process in metallurgy; it alters the physical and sometimes chemical properties of a material to increase its ductility and reduce its hardness, making it more workable. During the process atoms migrate in the crystal lattice and the number of dislocations decreases, leading to the change in ductility and hardness. In our experiment, we expect to decrease the number of defects by this process. Recently, Badawi et al. used an annealing process in order to study the behavior of the defects during annealing. They studied on different materials such as Mg, Al, Cu, Ag, Pb and Bi. Besides annealing, quenching is the rapid cooling of a sample to obtain certain material properties. During the quenching process, the extremely rapid cooling can prevent the formation of a crystal structure, thence resulting in an amorphous metal. Quenching experiments are usually performed with much thinner specimens, at least on the order of 1 mm in thickness to ensure a uniform quenching rate throughout the specimen. There are numerous complexities that can happen in quenching condition such as the formation of divacancies and higher order complexes, the production of dislocations and stacking faults rather than dislocations only, and vacancy loss from the specimen.

Our experimental setup is built on gamma ray detector and NIM module which is described in details in the experimental section in chapter 4. It is very encouraging that in 2009, Nam et al. used similar experimental setup for the defect study on polyaniline. They used $^{22}$Na as a positron source and BaF$_2$ scintillator with a photomultiplier tube for the experiment. The effect of perfect positioning of the scintillation detector might have great interest in question, however, it does not effect in the experimental data collection. This kind of understanding on positioning detector is very important in order to work with large specimen. They modified their spectrometer by using slanting scintillators and placed the photomultiplier tube right-angle to each other (Fig.1.2.2). The positron source was put with a reference material piece onto the surface of the specimen. After the first tests, they found that the results from this modified spectrometer and the results from collinear photomultipliers are the same.

All of these strong backgrounds on the system and corresponding theories and understanding have cogently persuaded us to study the source contribution in our sample with the system setup.

1.3 Advantages of this research

The determination of source contribution in the sample is extremely important in order to understand defect concentration in the sample. A pure sample could have different kinds of defect such as monovacancy, divacancies and different kinds of dislocation and so many things. By subtracting source contributing term from the spectrum the defect information can be extracted. By annealing and quenching process, it will be a detailed information about the behavior during those processes. It so much beneficial when it is applied to a pure metal and a commercial grade metal, because the commercial grade metal is an amalgamation of different alloys. Previously our group worked on steel, aluminum and copper. In that work, defects upon usage on the samples were studied through PALS technique, however, the source contributions were not calculated. Therefore, on the lifetime spectrum the contribution from the source is staying there and hence the solid information of the defects is questionable. In order to study any complex system such as commercial grade material of the corresponding atomic species, this technique can be applicable.
1.4 Reference


2 Structural properties of matter

2.1 Positron interaction

Positrons are elementary anti-particles that have half-integer spin and obey Fermi-Dirac Statistics. In the Standard Model, they are in the group of anti-leptons and are not subject to the strong nuclear interaction. Positrons are also known as anti-electrons or counter part of the electron and have the same amount of electric charge, mass and spin as electrons except the positive charge. Positrons are represented as $e^+$ and have mass of $511\text{keV}/c^2$, electric charge $+1e$ and spin $\frac{1}{2}$. They were first predicted by Paul Dirac [1], who proposed that electrons can have both positive charge and negative energy. In that paper he introduced an equation, now called Dirac equation, which is a unification of quantum mechanics, special relativity and the new concept of electron spin to explain the Zeeman effect. Though the paper did not explicitly predict the unknown particles, however, it gave a hint that electrons could have either positive or negative energy as solutions. The first experimental evidence of positrons was provided by Carl David Anderson through cloud-chamber experiments where he allowed cosmic rays to pass through a cloud chamber and a lead plate [2]. A magnet surrounded this apparatus, causing particle tracks to bend in different directions based on their electric charge. The trail left by each positron appeared on the photographic plate with a curvature matching the mass-to-charge ratio of an electron, but in a direction that showed its charge was positive. It was first noticed by Dmitri Skobeltyn when he was trying to detect gamma radiation from cosmic rays and found the particles that were acting like electrons, however, their trajectories curved in the opposite direction in an applied magnetic field [3]. In 1936, Anderson was awarded the Nobel Prize for his discovery and it was the first evidence of antimatter in nature. In the Standard Model, the second and third generation of anti-leptons are antimuon, $\mu^+$, and antitau, $\tau^+$, respectively.

Antimatter-matter reactions: In the Standard Model, every particle has its corresponding antiparticle. If the particles and anti-particles come in contact, then they annihilate each other and produce other particles. For example, when a positron interacts with an electron, they annihilate and produce two gamma rays.

$$\quad -1e^- + +1e^+ \rightarrow 0\gamma + 0\gamma$$

It is impossible to produce one gamma ray from the electron-positron pair in free space, because violate conservation of momentum. However, it can happen in the presence of a heavy nucleus that can recoil to conserve momentum.

Production of positrons: The production of positrons can take place in two ways: pair production and beta decay. Pair production is the process of creation of elementary particles and their antiparticles. Pair production will occur when the incoming particle has energy above a threshold, higher than the total rest mass energy of the two particles in order to conserve energy and momentum. In addition, angular momentum, electric charge and lepton number of the produced particles would be zero. The evidence of pair production was first observed by Patrick Blackett in a counter-controlled cloud chamber experiment where high energy photons dominantly interact with matter and produced electron-positron pairs [4]. This observation earned him a Nobel Prize in 1948.

$$\quad 0\gamma \rightarrow -1e^- + +1e^+$$

Here the energy of high energy photons (MeV scale or higher) is converted to the particles’ mass through Einstein’s equation, $E = m_e c^2$ where the symbols carry their usual meanings.
Another way of producing positrons is by beta decay. Beta decay is defined as a spontaneous nuclear disintegration process by which unstable atoms become more stable by dissipating excess energy and then undergoing a change of one unit of positive charge without any change in mass number. All beta decays are mediated by the weak nuclear force. There are three types of beta decay, called electron emission, positron emission, and electron capture. The electron emission is also known as $\beta^-$ decay.

$$^{14}_{6}C \rightarrow ^{14}_{7}N + e^- + \bar{\nu}_e$$

In this process, $^{14}_{6}$C becomes a new element with unchanged mass number, 14, but change in atomic number by one, this process is called nuclear transmutation. In the $\beta^-$, a neutron in an atom's nucleus turns into a proton, an electron and an antineutrino. The electron and antineutrino fly away from the nucleus, therefore, the nucleus gains a proton.

$$n \rightarrow p + e^- + \bar{\nu}_e$$

The antineutrino is an elementary particle with half integer spin, an anti-lepton which is the counterpart of the neutrino. In the Standard Model, the second and the third generation of antineutrino are muon antineutrino, $\bar{\nu}_\mu$ and tau antineutrino, $\bar{\nu}_\tau$ respectively.

Positron emission is known as $\beta^+$ decay and through this decay a positron is produced.

$$^{23}_{12}Mg \rightarrow ^{23}_{11}Na + e^+ + \nu_e$$

In this reaction, $^{23}_{12}$Mg turns into $^{23}_{11}$Na by emission of a positron and a neutrino where mass number does not change. However, the atomic number changes by one. In the $\beta^+$ decay, a proton in an atom's nucleus turns into a neutron while rejecting a positron and a neutrino.

$$p \rightarrow n + e^+ + \nu_e$$

The neutrino is an elementary particle (a lepton) with half integer spin, which interacts only via weak interaction and gravity. The second generation of neutrino is $\nu_\mu$ and the third generation is $\nu_\tau$ in the Standard Model.

Electron capture is the third form of Beta decay. Electron capture is the only type of decay that is allowed in proton-rich nuclides that do not have sufficient energy to emit a positron and neutrino$^5$.

$$p + e^- \rightarrow n + \nu_e$$

In this process, an atomic electron from the inner shell enters the nucleus and a proton changes to a neutron and simultaneously causes the emission of a neutrino. For example, $^7$Be turns into $^7$Li by emission of a neutrino.

$$^7_{3}Be + e^- \rightarrow ^7_{3}Li + \nu_e$$

For our experiment, we use $^{22}_{11}$Na, a cosmogenic isotope, which decays into $^{22}_{10}$Ne. The energy level diagram and decay scheme is shown in Fig. 2.1.1. It occurs by one of two mechanisms: positron decay or electron capture.

$$^{22}_{11}Na \rightarrow ^{22}_{10}Ne (ground \ space) + e^+ + \nu_e \ (0.06\% \ of \ all \ decays)$$
and

\[
^{22}_{11}\text{Na} \rightarrow ^{22}_{10}\text{Ne}^* (1.275\text{MeV}) + e^+ + \nu_e \quad (90.4\% \text{ of all decays})
\]

The ground state of \(^{22}\text{Ne}\) is stable and the first excited state of \(^{22}\text{Ne}\) at 1.275MeV decays with a lifetime 3.7ps by gamma decay process;

\[
^{22}_{10}\text{Ne}^* (1.275\text{MeV}) \rightarrow ^{22}_{10}\text{Ne} (\text{ground state}) + \gamma_{1.275\text{MeV}}
\]

which gives rise to a characteristic gamma ray with energy 1.275 MeV. The positrons eventually disappear in the annihilation process and produces two characteristic 0.511 MeV annihilation gamma rays.

\[
e^- + e^+ \rightarrow 2\gamma_{0.511\text{MeV}}
\]

In addition, electron capture occurs in about 9.5% of all decays.

\[
^{22}_{11}\text{Na} + e^- \rightarrow ^{22}_{10}\text{Ne}^* (1.275\text{MeV}) + \nu_e (9.5\% \text{ of all decays})
\]

The electron capture process populates only the first excited state of \(^{22}\text{Ne}\) at 1.275 MeV and therefore the characteristic 1.275 MeV gamma rays result. The annihilation of gamma rays at 0.511 MeV are not produced in electron capture, because positrons are not created.

In solids, the trapping of positrons in defects is based on the formation of an attractive potential that is caused by lack of a repulsive positively charged nucleus at open-volume defects, such as vacancies, vacancy agglomerates, and dislocations. The sensitivity range for the vacancy detection in metals starts at one vacancy per \(10^7\) atoms. This enormous sensitivity is caused by the fact that in the course of its lifetime the positron diffuses about 100nm through the lattice and thus probes a high number of atoms until annihilation. When the positron is trapped in an open-volume defect, the annihilation parameters are changed in a characteristics way. The positron lifetime increases in an open-volume defect, because the electronic density in the region of vacancy is lower than in the bulk. In the clustering of vacancies, the positron lifetime increases more due to the lower electron density. Positrons trap in different types of defects have different lifetimes.

### 2.2 Structural properties of aluminum

Aluminium is the most abundant and most widely used metal after steel. It is the third most common element comprising 8.2% of the earth’s crust after oxygen and silicon. Aluminium is so chemically reactive that native specimens are rare and limited to extreme reducing environments. Instead, it is found combined in over 270 different minerals\(^6\). It is never found free in nature. The chief ore of aluminium is bauxite. The name “aluminum” was derived from the ancient name for alum (potassium aluminium sulphate), which was alumen (i.e.: Latin, meaning bitter salt). Aluminum was the original name given to the element by Sir Humphry Davy in 1807, but later scientists respelled it by a pleasant name “aluminium” and that became the accepted name in Europe. However, in the USA the preferred name is aluminum that was accepted after the meeting of American Chemical Society in 1925.
In the periodic table, aluminum is in the boron group with symbol “Al” and atomic number “13”. It is remarkable for the metal’s low density and its ability to resist corrosion through the phenomenon of passivation. In our lifestyles and built environment, aluminum products are just as abundant. Since its commercial production began little more than a century ago, aluminum has become the material of choice for a diverse range of applications and utilities. In addition, it is possible to recycle and resell a discarded aluminum within very short time period such as 60 days. Its elegance and versatility gave this metal a unique reputation that once aluminum was considered to be a precious metal and even more valuable than gold. It is said that once the Emperor of France, Napoleon III gave a banquet where the most honored guests were given aluminum cutlery, while everyone else had gold.

General properties of aluminum:
Aluminum belongs to the class of white metals, and has a high luster. When pure it is as white as silver, but the commercial metal has a bluish tinge, due to the presence of impurities. It is slightly harder than tin, and its hardness is increased by the presence of impurities. The fracture of the ingot is uneven but somewhat close grained, that the worked metal has a finer grain, and is silky in appearance.

Density:
Aluminum is a very light metal with a specific weight of 2.7 g/cm³, about a third that of steel. For example, the use of aluminum in vehicles reduces dead-weight and energy consumption while increasing load capacity. Its strength can be adapted to the application required by modifying the composition of its alloys.

Malleability and ductility:
Aluminum is ductile and has a low melting point, 660.32°C as well as density. In a molten condition, there are number of ways that its processing can be done. Its ductility allows products of aluminum to be basically formed close to the end of the product’s design. It can be forged and rolled and even be beaten into thin leaves in the frequent annealing condition. It can be drawn into tubes without any difficulty, and also into fine wire. It is very malleable between 100°C and 150°C. The metal can be spun, stamped, and pressed. It can also be extruded in the form of rods or tubes, at a temperature of about 80°C, by hydraulic pressure with suitable dices.

Tensile strength:
Pure aluminum doesn’t have a high tensile strength, however, it can be increased by adding alloy elements like manganese (Mn), silicon (Si), copper (Cu) and magnesium (Mg), and that can be produced an alloy with properties tailored to particular applications.

<table>
<thead>
<tr>
<th>Atomic number</th>
<th>13</th>
</tr>
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<tbody>
<tr>
<td>Electron configuration</td>
<td>[Ne]3s²3p¹</td>
</tr>
<tr>
<td>Group, block</td>
<td>group 13 (boron group), p-block</td>
</tr>
<tr>
<td>Period</td>
<td>period 3</td>
</tr>
<tr>
<td>Element category</td>
<td>post-transition metal</td>
</tr>
<tr>
<td>Atomic mass</td>
<td>26.98 g/mol⁻¹</td>
</tr>
<tr>
<td>Density</td>
<td>2.7 g/cm³</td>
</tr>
<tr>
<td>Melting point</td>
<td>660.32°C</td>
</tr>
<tr>
<td>Boiling point</td>
<td>2470°C</td>
</tr>
<tr>
<td>Vanderwaals radius</td>
<td>184 pm</td>
</tr>
<tr>
<td>Covalent radius</td>
<td>121 ± 4 pm</td>
</tr>
<tr>
<td>Energy of 1st ionization</td>
<td>577.5 kJ/mol⁻¹</td>
</tr>
<tr>
<td>Energy of 2nd ionization</td>
<td>1816.7 kJ/mol⁻¹</td>
</tr>
<tr>
<td>Energy of 3rd ionization</td>
<td>2744.8 kJ/mol⁻¹</td>
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<tr>
<td>Energy of 4th ionization</td>
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<tr>
<td>Energy of 5th ionization</td>
<td>14842 kJ/mol⁻¹</td>
</tr>
<tr>
<td>Prediction</td>
<td>Antoine Lavoisier</td>
</tr>
<tr>
<td>Named by</td>
<td>Sir Humphry Davy</td>
</tr>
</tbody>
</table>

Table 2.2.1: General properties of aluminum
Aluminum is well suited to cold environments. It has the advantage over steel in that its’ tensile strength increases with decreasing temperature while retaining its toughness. Steel on the other hand becomes brittle at low temperatures. When aluminum is cast in sand, its tenacity is 5 tons per square inch, with an elongation of 25% when it is in chills, then 5.25 tons per square inch with an elongation of 35%, and while rolled bars and hard drawn wire have respectively tenacities of 7 and 15 tons per square inch.

**Action of heat:**
The melting point of aluminum is 660.32°C. It volatilizes at a very high temperature, however, the volatilization point is not exactly determined.

**Chemical properties:**
When aluminum is exposed to air, it forms almost instantaneously a thin surface layer of aluminum oxide that effectively prevents further oxidation, in a process called passivation. This layer has excellent resistance to corrosion and it is fairly resistant to most acids but less resistant to alkalis. The strongest aluminum alloys are less corrosion resistant due to galvanic reactions with alloyed copper. In the presence of aqueous salt, the corrosion resistance can be reduced, particularly with the dissimilar metals.

At temperature below 280°C, aluminum is oxidized by water and produced aluminum hydroxide and hydrogen.

\[ 2Al + 6H_2O \rightarrow 2Al(OH)_3 + 3H_2 \]

This conversion is made of interest for the production of hydrogen. However, commercial application of this fact has challenges in circumventing the passivating oxide layer, which inhibits the reaction, and in storing the energy required to regenerate the aluminum metal.

**Health effect:**
Aluminum is commonly known as innocent compound. However, it can be harmful for health with exposed to its high concentration. The water-soluble form of aluminum usually found in a solution of aluminum causes the harmful effects.

Long lasting exposure to aluminum and uptake through breathing and by skin contact has serious health problems such as Alzheimer, dementia, listlessness, loss of memory, damaging to the central nervous system and Severe trembling. It is also risky in mining work as it can be found in water. People that work in factories where aluminum is applied during production processes may endure lung problems when they breathe in aluminum dust. Aluminum can cause problems for kidney patients when it enters the body during kidney dialyses.

### 2.3 Structural properties of lead

Lead is one of the widely used metals since its discovery of more than 6000 years ago. Its elegance of easy casting, shaping and joining lead articles and resistant to corrosion have made it vastly used material over century and centuries. There are many examples of lead articles which have lasted for centuries. Lead is relatively abundant occurs naturally in the Earth's crust, and it can be easily extracted from the ore of lead concentration and winning the metal from the concentrate does not need much energy. This reflects in a fairly low price compared with other non-ferrous metals. Lead can be recycled as a secondary raw material from lead-acid batteries, from metallic scrap and from several composite consumer products in conjunction with existing recycling loops, for example for steel, zinc and copper, at moderate costs.

However, lead has extremely low strength compare to other metals that is exacerbated by its creep and fatigue behavior. So it is unsuitable for applications that require even moderate strength. However, small
alloying additions can increase its strength considerably. Thus it can be used those applications that require higher strength like lead clad steel.

Lead can be used different kinds of applications because of its high density such as shielding against sound, vibrations and radiation, for example as protection for users of computer and TV screens. Some compounds of lead have their own useful properties, particularly in relation to color and glass-forming ability.

General properties of lead:
Freshly casted lead is silver in color, however, soon it turns dull grey to bluish grey in the presence of air due to the formation of surface oxide. The liquid metal has shiny chrome-silver luster. A cast ingot of lead is generally called a ‘pig’ that yields a very dull sound when struck with hammer. As much as duller the sound that much as purer the metals. The approximate purity also can be determined by its hardness when it is scratched with finger nails\[11\].

Lead possesses the physical properties of metals such as electrical and thermal conductivity, metallic luster, and high density. It has low melting point compared with other metals, 327.5°C.

There are four isotopes of lead found in nature; $^{204}$Pb, $^{206}$Pb, $^{207}$Pb and $^{208}$Pb – those are stable and have magic number with 82 protons in each. One of the isotopes, lead-208 has 126 neutrons that is another magic number, hence more stable. Lead-208 is the heaviest known stable nuclide. The isotopes and their relative concentrations in different mineral deposits can vary; this can be used as a diagnostic tool in identifying sources of lead in soils and sediments. The isotope $^{210}$Pb can be occurred in nature in extremely small amount, the so called trace isotope. And the isotope, $^{206}$Pb does not happen in nature and thence called synthetic isotope.

In the Periodic table, lead is a heavy post-transition metal and it is the highest atomic number element with non-radioactivity (the next element bismuth has radioactive isotope with very long half-life that considered to be stable and the other two radioactive elements; Technetium and Promethium are lighter).

Density:
Lead has comparatively higher density with other metals. Its density is 11.3g/cm$^3$ whereas iron, copper and aluminum have 7.8g/cm$^3$, 8.9g/cm$^3$ and 2.7g/cm$^3$ respectively. Lead owes its high density to two factors:
1. It has high atomic number, 82 and hence high relative atomic mass of 207.
2. The metallic structure of lead is closest packed structures which is the most tightly packed or space-efficient composition of crystal structures.

Table 2.3.1: General properties of lead

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atomic number</td>
<td>82</td>
</tr>
<tr>
<td>Electron configuration</td>
<td>[Xe]$4f^{14}5d^{10}6s^26p^2$</td>
</tr>
<tr>
<td>Group, block</td>
<td>group 14 (carbon group), p-block</td>
</tr>
<tr>
<td>Period</td>
<td>period 6</td>
</tr>
<tr>
<td>Element category</td>
<td>post-transition metal</td>
</tr>
<tr>
<td>Atomic mass</td>
<td>207.2 g/mol$^{-1}$</td>
</tr>
<tr>
<td>Density</td>
<td>11.34 g/cm$^3$ at 20°C</td>
</tr>
<tr>
<td>Melting point</td>
<td>327.5°C</td>
</tr>
<tr>
<td>Boiling point</td>
<td>1755°C</td>
</tr>
<tr>
<td>Vanderwaals radius</td>
<td>0.154 nm</td>
</tr>
<tr>
<td>Ionic radius</td>
<td>0.132 nm (+2); 0.084 nm (+4)</td>
</tr>
<tr>
<td>Energy of 1$^{\text{st}}$ ionization</td>
<td>715.4 kJ/mol$^{-1}$</td>
</tr>
<tr>
<td>Energy of 2$^{\text{nd}}$ ionization</td>
<td>1450 kJ/mol$^{-1}$</td>
</tr>
<tr>
<td>Energy of 3$^{\text{rd}}$ ionization</td>
<td>3080.7 kJ/mol$^{-1}$</td>
</tr>
<tr>
<td>Energy of 4$^{\text{th}}$ ionization</td>
<td>4082.3 kJ/mol$^{-1}$</td>
</tr>
<tr>
<td>Energy of 5$^{\text{th}}$ ionization</td>
<td>6608 kJ/mol$^{-1}$</td>
</tr>
<tr>
<td>Discovered by</td>
<td>The ancients</td>
</tr>
</tbody>
</table>
3. This structure is also called face-center cubic (fcc) structure.

The high atomic mass means that more electrons are needed to maintain a neutral charge and its face-centered cubic structure allows it to have small bond length and a small atomic radius means that many atoms can be packed into a particular lead structure. Therefore, its density and large number of electrons make it well-suited for scattering X-rays and gamma rays, that is, the photons of these rays impart energy onto electrons when they come into contact. On the other hand, it has high positively charged nucleus, so the electrons around the middle of it can absorb a huge amount of energy before they get kicked off the atom. Hence, an electron which is very near to the center of the nucleus, can absorb a much more energetic X-ray and gamma ray.

However, it needs to be pointed out that some other elements have higher density than lead such as gold; 19.3g/cm$^3$, platinum; 21.5g/cm$^3$, tungsten; 19.3g/cm$^3$, tantalum; 16.6g/cm$^3$ and mercury; 13.6g/cm$^3$. There are three metals with similar densities to lead are molybdenum; 10.2g/cm$^3$, silver; 10.5g/cm$^3$ and palladium; 12.0g/cm$^3$. However, lead is used as radiation shielding as it is cheaper and abundant in nature than those metals.

**Attenuation of X-rays and gamma rays:**

When gamma radiation of intensity, $I_0$ is incident on an absorber of thickness, $x$ and the emerging intensity, $I_x$ transmitted by the absorber is related by following equation,

$$-dl \propto ldx = \mu ldx$$

Here $\mu$ is a proportionality constant, called linear attenuation coefficient.

Dividing across by $l$, and thus integrating over gamma ray transmission, $dl/l$ over thickness, $x$,

$$-\frac{dl}{l} = \mu dx$$

$$-\int_{I_0}^{I_x} \frac{dl}{l} = \mu \int_0^x dx$$

$$\frac{I_0}{I_x} = I_0 e^{-\mu x}$$

This final expression tells that the radiation intensity will decrease in an exponential fashion with the thickness of the absorber with the rate of decrease being controlled by the Linear Attenuation Coefficient. Lead has a high mass attenuation coefficient, particularly for higher energy X-rays (as have other elements of high atomic number) and high density, so it makes an excellent shielding material.

**Mechanical properties of lead:**

Under low loads, all solid materials deform slightly, then return to their original shape when the load is released, that is called elastic deformation and it is reversible. However, in case of higher stress, brittle
materials fracture and ductile ones (some metals, some polymers) are plastically deformed, i.e. can be stretched, bent etc. and do not return to their original shape when the load is released.

Malleability and ductility:

Lead has low ductility, but high malleability\textsuperscript{[12]}. It can be readily rolled into sheets and even into thin foil. However, because of its low ductility it cannot be drawn into fine wire. When the metal is heated to a certain temperature it may be squeezed through an aperture into pipes and solid rods. This operation is called squirting and all lead pipes are made by this operation.

Tensile strength:

The tensile strength of pure lead is about $12 - 17\ MPa$, which is very much lower than that of the other common metals such as mild steel is about 15 times stronger, cast copper is 10 times stronger, cast aluminium is about 6 times stronger. Even less strong materials such as soft pine, plastics have slightly higher strengths than lead. However, as lead slowly deforms with time under a static load and this phenomenon is called creep and hence it is necessary to specify the exact testing conditions.

Table 2.3.2: Comparison of the tensile strength of some common materials with lead

<table>
<thead>
<tr>
<th>Metals</th>
<th>Pb</th>
<th>Cu</th>
<th>Fe</th>
<th>Al</th>
<th>Zn (rolled)</th>
<th>Sn</th>
<th>Soft solder</th>
</tr>
</thead>
<tbody>
<tr>
<td>UTS (MPa)</td>
<td>12 -17</td>
<td>120 – 170</td>
<td>100 – 230</td>
<td>90 – 100</td>
<td>110 – 150</td>
<td>20 – 35</td>
<td>55 – 75</td>
</tr>
</tbody>
</table>

For metals, strength depends on temperature, material preparation, rate of cooling, any deformation or further heat treatment, and the presence of impurities even in trace amounts. For glass, the amount of handling has a marked effect in decreasing strength and in case of polymers, they are sensitive to variables in manufacturing process, and degrade after exposure to heat and ultra-violet radiation\textsuperscript{[13]}.

Though the tensile strength of lead is very low, however, it can be increased by adding alloy. The effect of adding of antimony with lead is given in table 2.3.3.
Table 2.3.3: Strengthening Effect of adding Antimony to Lead[14]

<table>
<thead>
<tr>
<th>Antimony content (in %)</th>
<th>Tensile strength (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>17.25</td>
</tr>
<tr>
<td>1</td>
<td>23.46</td>
</tr>
<tr>
<td>2</td>
<td>29.00</td>
</tr>
<tr>
<td>4</td>
<td>39.05</td>
</tr>
<tr>
<td>6</td>
<td>47.20</td>
</tr>
<tr>
<td>8</td>
<td>51.20</td>
</tr>
<tr>
<td>10</td>
<td>52.92</td>
</tr>
<tr>
<td>11</td>
<td>52.58</td>
</tr>
<tr>
<td>12</td>
<td>57.61</td>
</tr>
</tbody>
</table>

Action of heat:
The melting temperature of lead is 327.5°C and when the pure lead is heated to its melting point and then broken. The fractured surface show largely columnar structure and larger column the purer the lead. If the air be excluded, lead is not sensibly volatized below a red heat, and it cannot be distilled like zinc. However, the distillation of zinc or zinc ores contain lead, and a considerable amount is carried over by the zinc vapor. With access of air fumes of lead oxide are evolved at a bright red heat which can be used as smelting operations.

Chemical properties:
In perfectly dry air lead undergoes not change at ordinary temperature, but it becomes dull from oxidation due to moist air. At its melting point, lead oxide, $\text{PbO}$, is formed, and at high temperature, the oxidation takes place rapidly.

Lead oxide, $\text{PbO}$ is also called lead mono-oxide. Commercially it is called litharge. The oxidizing action of litharge plays an important part in the treatment and assaying silver and its ores. When it is fused with S, Fe, As, Sb, Bi, Sn, Cu, and Zn, then these substances become wholly or partially oxidized, and their oxides either volatilize, or dissolve in the excess of litharge. The other oxides except red lead, $\text{Pb}_3\text{O}_4$, are of little importance in metallurgy, and $\text{Pb}_3\text{O}_4$ only in assaying.

Health effect:
Lead and its compounds are poisonous to animals and humans if ingested or inhaled. Lead is a neurotoxin that accumulates both in soft tissues and the bones, damaging the nervous system and causing brain disorders. Excessive lead also causes blood disorders in mammals. Lead poisoning has been documented since ancient Rome, ancient Greece, and ancient China.

Lead compounds are used as a pigment in paints, dyes, and ceramic glazes and in caulk. The amount of lead used in these products has been reduced in recent years to minimize lead’s harmful effect on people and animals. Tetraethyl lead and tetramethyl lead were once used in the United States as gasoline
additives to increase octane rating. However, their use was phased out in the United States in the 1980s, and lead was banned for use in gasoline for motor vehicles beginning January 1, 1996.

Most lead used by industry comes from mined ores ("primary") or from recycled scrap metal or batteries ("secondary"). Lead is mined in the United States, primarily in Alaska and Missouri. However, most lead today is “secondary” lead obtained from lead-acid batteries. It is reported that 97% of these batteries are recycled.

Lead comes to environmental effect through human activities. Environmental levels of lead have increased more than 1,000-fold over the past three centuries as a result of human activity. The greatest increase occurred between the years 1950 and 2000, and reflected increasing worldwide use of leaded gasoline. In 1979, cars released 94.6 million kilograms (208.1 million pounds) of lead into the air in the United States. In 1989, when the use of lead was limited but not banned, cars released only 2.2 million kg (4.8 million pounds) to the air. Once lead gets into the atmosphere, it may travel long distances if the lead particles are very small. Lead is removed from the air by rain and by particles falling to land or into surface water.
2.4 Reference

3 Theory of positron annihilation lifetime spectroscopy

3.1 Defects in materials

The appearance of defects in materials might be considered as a blessing for discovery and versatile applications. Defects are so interesting as well as extensively broader in application that they can modify the properties of a solid which can later develop with lots of versatility with engineering and technology and then come up with useful applications. The evolutionary history of the understanding of defects has gradually developed. Most probably the earliest concept of defects in a solid is a wrong atom, that is an impurity and thus a so-called point defect. It might be the simplest defect found in a solid. Defects in a crystal lattice can be classified by their dimension,

1. Zero-dimensional defects – Point defects
2. One-dimensional or linear defects – dislocations
3. Two-dimensional or planar defects – external and internal surfaces
4. Three-dimensional or volume defects – point defect clusters, voids, precipitates.

These defects are called structural defects. In addition to these, there are also electronic defects. The electronic defect has two varieties such as current is carried by excess electrons and other type is current is carried by particles which is called electron holes, positive holes or just simply holes.

It is quite evident that by default, defects have come to be associated with crystalline solids because of regular repetition of atoms in a crystal\(^1\). Therefore, the defects occur depending on the disturbance of the regularity.

3.1.1 Point defect

The simplest type of defect in a crystal might be defined as the mistake at a single atom site in a pure monoatomic crystal. Simply two types of point defects can occur in a pure monoatomic crystal – vacancy type point defect and the other is interstitial point defect. Vacancy type defects happen during crystal growth and it can be defined as missing atoms in regular occupied position that leaves a vacancy. The interstitial defects occur during crystallization by interstitial atoms that happens when an extra atom is forced to take position in an interstitial site that is normally unoccupied position in the crystal lattice. If the extra atom is same spices of the normal atoms, then it is called self-interstitial atom.

In addition to these type of point defects, there are other types of point defects that are really of great interest such as native defects and induced defects. Native defects occur in a crystal as grown such as vacancies, interstitials, excess electrons, holes or other arrangements. Although native defects can be introduced during crystal growth, there are several ways of forming native defects. Induced defects are happening when the crystal is subject to irradiation by high-energy particles or other form of radiation. The number of native defects can be changed by an annealing process, however, the population of point defects will always remain the same even if that is the purest crystal\(^1\). These point defects are in thermodynamic equilibrium and they cannot be removed from the solid they are so called intrinsic point defects. Thus the residual population of the defects are temperature dependent and their number will be increased with progressive temperature. Intrinsic point defects can be more complex in ordered compounds. One of the simplest binary ionic solid is NaCl. In a NaCl crystal, the excess charge of Na, that is cation, thus vacancy is balanced if there is a Cl vacancy nearby which is anion. Therefore, A neutral defect that involves paired vacancies on the cation and anion sublattices is called a Schottky defect. Alternatively, a neutral defect that is made up of a paired vacancy and interstitial is called a Frenkel defect. For example, in a NaCl crystal, it could happen when the Na vacancy is compensated by a Na interstitial. In addition to these, anti-site defects could happen in the compound. This occurs mainly.
when the compound atoms are less strongly ionized, then it is energetically possible for the species to exchange their sites. This kind of defect is mostly common in semiconductors such as GaAs.

The opposite of intrinsic type of point defect is extrinsic point defects; these are necessary for semiconductor engineering. Extrinsic point defects occur when a foreign atom is added in the crystal. When the foreign atom is added in the crystal intentionally, then that is called solutes, and otherwise, it is called impurities. If it happens in the lattice site, then that is called substitutional solutes, or in contrast, substitutional impurities. However, if that happens in the interstitial site, then it is called interstitial solute, and similarly, the opposite one is interstitial impurities. Typically, interstitial sites are very small compared with substitutional. Small atoms such as hydrogen, carbon, nitrogen are often found in the interstitial site, and larger atoms are usually substitutional. Generally, complex extrinsic defects are found in compounds. In the semiconductor industry, the point defects are intentionally added in order to control type and concentration of the charge carrier. Depending on the type of the dopant, (i.e. the foreign atom that is added), there are two types of defects – p-type impurity, thus so called p-type semiconductor, and similarly, the other type is n-type semiconductor.

3.1.2 Dislocations

Dislocations are linear defects which enable us to understand the strength of a metal. Typically, a dislocation is associated with deformation of the metal in a sense that a dislocation free metal requires less energy to deform. If the dislocation movement is impeded or impossible, then the material would be hard and brittle. For example, ceramics which are brittle and they are blocking dislocation movement rather than have a lack of dislocations. At high temperature, ceramics act like metals at low temperature, because the dislocation movement is increased. There are different types of defects due to dislocations, such as edge dislocations, screw dislocations and other types with different combinations. For our experiment, we are not concerned with different types of dislocations or other higher order defects. We are trying to understand closely the behavior of intrinsic point defects in metals, thence, the other types of defects are not necessarily important to describe here.

3.1.3 Diffusion in solids and defects

Diffusion is a process by which atoms, molecules or ions move in a surrounding medium. It is an important factor in order to understand reaction of solids with other solids, liquids or gases. In a different context, it is something that is very useful to describe the behavior of species that are travelling around in the corresponding medium. It certainly depends on the concentration of diffusing species at different distances from the release point within a time period at a certain temperature.

Therefore, mathematically it can be presented by Fick’s first and second law,

Fick’s first law\(^{[1]}\)

\[
J = -D \frac{dc}{dx}
\]  \hspace{1cm} (1)

where \(J\) is the flux of diffusing species, \(D\) is diffusion coefficient and \(c\) is the concentration of diffusing species.

Fick’s second law\(^{[1]}\)

\[
\frac{dc}{dt} = -D \frac{d^2 c}{dx^2}
\]  \hspace{1cm} (2)
Here $x$ and $t$ are for distance and time, respectively.

In solids, the diffusion is significantly depending on the structure of the matrix across which diffusion occurs and the presence of defects. In simple crystalline solids, diffusion is mediated by the defects and the diffusion rate changes with the type of defects in that solids. Diffusion movement through the body of a solid is called volume, lattice or bulk diffusion. In a cubic crystal, the bulk diffusion is same in all directions. It is the same in amorphous or glassy solids. In gases and liquids, the diffusion movement is also the same in all directions and the material is described as isotropic. In all other crystals, diffusion depends on the direction, and the material is treated as anisotropic. For a perfect crystal, point defects play a significant role for bulk diffusion. In case of higher dimensional defects such as two dimensional defects (dislocations), the diffusion is much faster than bulk diffusion at lower temperature. This process is called short-circuit diffusion.

The most popular, simplest and basic diffusion model is called random-walk diffusion model; it states that diffusion of atoms across the bulk of a solid follow in such a way that they move by a series of random jumps, because all the atoms are being continually jostled by thermal energy. Hence they follow a random walk. It is also called drunkard’s walk\(^1\).

According to the one-dimensional random-walk model, if $N$ is the number of diffusing atoms that are carrying out $n$ random steps over the period of time, $t$, then the average of the square of the distance for each atom, $\langle x \rangle^2$ is given by\(^1\),

$$\langle x \rangle^2 = na^2 = \Gamma t a^2$$  \hspace{1cm} (3)

Here $a$ is the distance between two jumps and $\Gamma$ is the jumping frequency of the atoms from one position to the next position.

However, it is significantly interesting that net atom displacement does not depend on jumps rather random movement alone. Therefore, in terms of diffusion coefficient, $D$, equation (3) can be written as,

$$\langle x \rangle^2 = 2D_r t$$  \hspace{1cm} (4)

Here $D_r$ is random walk diffusion coefficient and the factor 2 arises for geometrical reasons in one dimension. Equation (4) is called Einstein-Smoluchowski Diffusion equation or simply Einstein diffusion equation\(^1\).

In case of two and three dimensions, the above equation is given by,

In two dimension, 

$$\langle x \rangle^2 = 4D_r t$$

In three dimension, 

$$\langle x \rangle^2 = 6D_r t$$

Thus, the shape of the penetration profile due to random-walk of the diffusion can be derived as\(^1\),

$$N(x) = \frac{2N}{\sqrt{2\pi n}} e^{\left(-\frac{x^2}{4D_r t}\right)}$$  \hspace{1cm} (5)

where $N(x)$ is the number of atoms that reach position $x$. 

20
Equation (5) represents a bell shaped curve, therefore, by statistical interpretation, the penetration depth for one-dimensional diffusion is given by,

\[ x_p \approx \sqrt{2Dt} \quad (6) \]

where \( D \) is the appropriate diffusion coefficient of time \( t \).

The equation (6) sometimes can, arbitrarily, be written as\(^{[1]}\),

\[ x_p \approx 2\sqrt{2Dt} \quad (6.1) \]

Hence, diffusion length is,

\[ L = \sqrt{Dt} \quad (7) \]

The random walk model can explain vacancy diffusion in solids. Thus if there is a concentration gradient of a species (e.g.: vacancies) exists if there is population of defects such as vacancies. For vacancies, atoms jump from a normal site into neighboring vacancy and so move through the crystal. Similarly, for the movement of vacancy, it moves one site to another site as if it is a particle. This type of movement of the vacancies is called vacancy diffusion. It is also known as indirect diffusion. It is a common phenomenon in pure metals.

Similarly, the diffusion of the interstitial site occurs by a jump of an interstitial to a neighboring interstitial. This type of diffusion is called interstitial diffusion. Sometimes it is treated as direct diffusion.

In addition to these, temperature has considerable effect on diffusion coefficient which can be given by an Arrhenius equation\(^{[2]}\),

\[ D = D_0 e\left(\frac{-E_a}{RT}\right) \quad (8) \]

Here, \( D_0 \) is a constant. It is sometimes treated as pre-exponential factor, frequency factor and more occasionally diffusion coefficient, \( R \) and \( T \) are gas constant and temperature in Kelvin respectively.

However, there is a serious weakness of the random walk model which does not allow temperature variation. Thus it requires a modification to take temperature variation into account.

The modified equation is given as\(^{[2]}\),

\[ D = \gamma f[d]a^2v e\left(\frac{-\Delta H_m}{RT}\right) \quad (9) \]

where \( \gamma \) is a combined geometrical and entropy factor, \( f \) is the correlation factor which is given by equation (10), \( [d] \) is defect concentration involved in diffusion, \( a \) is the jump distance from one site to next site during diffusion, \( v \) is the attempt frequency, and \(-\Delta H_m\) is the molar enthalpy of migration.

Correlation factor,

\[ f = \frac{\langle x \rangle_{correlated}^2}{\langle x \rangle_{random}^2} \quad (10) \]

Here \( \langle x \rangle_{correlated}^2 \) stands for the mean square displacement of the atom which is moving in a correlated motion and \( \langle x \rangle_{random}^2 \) is the mean square displacement of the atom or vacancy moving by a random walk.
3.2 Trapping model

The trapping model is frequently used for analyzing experiments on positron annihilation in solids. It is also called two state trapping model, or Brandt-Bergersen-Stott-Connors-West trapping model. It assumes that the positron exists in one of only two states in the metal, the bulk or Bloch state and the defect-trapped state.

Let us consider a metal in which the positrons will be injected. The positrons are emitted from a radioactive source and those become thermalize rapidly (~1ps) which results in the positrons that are initially in a free state sometimes also referred to as Bloch-like state.

Bloch-like state in the lattice. Those positrons can subsequently be trapped in a highly localized state in a defect. Therefore, upon interaction with electrons, positrons annihilate and each positron-electron pair emits two 511keV γ-rays.

Mathematically, the trapping equations can be described as follows[3],

\[
\frac{dn_b}{dt} = -n_b \lambda_b - n_b k_t \\
\frac{dn_t}{dt} = +n_b k_t - n_t \lambda_t
\]  

Here \(n_b\) and \(n_t\) denote fraction of free (untrapped) and defect-trapped positrons, respectively, with \(n_b(0) = 0\), \(\lambda_b\) and \(\lambda_t\) are the positron-annihilation rates in the bulk and defect-trapped states, respectively, and \(k_t\) is the total transition or trapping rate of the positron from the bulk state to its defect-trapped state[2-3].

The positron lifetimes in the two states; \(\tau_b\) is for bulk state and \(\tau_t\) is for trapped state, are defined by,

\[
\tau_b = \frac{1}{\lambda_b} \\
\tau_t = \frac{1}{\lambda_t}
\]

Thus the fraction of positrons that are still “alive”[2],

\[
n(t) = n_b(t) + n_t(t)
\]

The solution of the coupled set of trapping equations (1) is of the form,

\[
n(t) = l_1 e^{-t/\tau_1} + l_2 e^{-t/\tau_2}
\]

With the intensities \(l_i\) of lifetime component \(\tau_i\) such that \(l_1 + l_2 = 1\). Lifetime \(\tau_1\) is associated with the positron lifetime in the free state, and lifetime \(\tau_2\) related to the positron lifetime in the trapped state.

The \(l_i\) and \(\tau_i\) are determined by least-square fitting the lifetime spectrum. Now we need to determine how the \(l_i\) and \(\tau_i\) are related to the quantities in equation (1),
The first equation (1) is a first order homogeneous differential equation,

\[ \frac{dn_b}{dt} + (\lambda_b + k_t)n_b = 0 \]  

and has the solution,

\[ n_b(t) = e^{-(\lambda_b + k_t)t} \]  

Putting this solution into the second equation (1) results in an inhomogeneous equation,

\[ \frac{dn_t}{dt} + n_t\lambda_t = k_t e^{-(\lambda_b + k_t)t} \]  

with the solution,

\[ n_t(t) = \frac{k_t}{\lambda_b - \lambda_t + k_t} (e^{-\lambda_t t} - e^{-(\lambda_b + k_t)t}) \]  

Combining equation (7) and (9) with equation (4), and using \( I_1 = 1 - I_2 \) yields,

\[ n_t(t) = \left(1 - \frac{k_t}{\lambda_b - \lambda_t + k_t}\right) e^{-(\lambda_b + k_t)t} + \frac{k_t}{\lambda_b - \lambda_t + k_t} e^{-\lambda_t t} \]  

From comparison of equation (5) and (10) we have,

\[ \tau_1 = \frac{1}{\lambda_b + k_t} \]  

\[ \tau_2 = \frac{1}{\lambda_t} \]  

and

\[ I_1 = \frac{\lambda_b - \lambda_t}{\lambda_b - \lambda_t + k_t} \]  

\[ I_2 = \frac{k_t}{\lambda_b - \lambda_t + k_t} \]  

From equation (11) we find that the annihilation rate is the free state is,

\[ \lambda_b = \frac{1}{\tau_1} - k_t \]  

and the annihilation rate in the trapped state is simply the inverse of \( \tau_2 \),

\[ \tau_2 = \frac{1}{\lambda_t} \]  

Solving equation (14) for \( \lambda_b \) and setting \( \lambda_b + k_t = \frac{1}{\tau_1} \) we find,

\[ k_t = I_2 \left(\frac{1}{\tau_1} - \frac{1}{\tau_2}\right) \]
The lifetime spectrum can also be characterized by the mean lifetime define by,

\[ \tau_m = \int_0^\infty n(t) \, dt \]  

(18)

Putting equation (5) into equation (14) we have,

\[ \tau_m = l_1 \tau_1 + l_2 \tau_2 \]  

(19)

If the two state trapping model describes the lifetime data well, then the mean lifetime \( \tau_m \) is between \( \tau_b = \frac{1}{\lambda_b} \) and \( \tau_t = \frac{1}{\lambda_t} \).

\[ \tau_b \leq \tau_m \leq \tau_t \]  

(20)

3.3 Positronium formation

In addition to the direct decay of positrons in the bulk state and defect trapped state, there is another mode of decay which is typically formed on the surface of the sample\(^4\), that is an electron-positron bound state called positronium (Ps). Positronium is the lightest know atom quite similar to a hydrogen atom. The thermalized positron cannot form Ps with one of the electrons bound in the molecules due to their high binding energy, \( 9 - 12 \, eV \). Typically, the binding energy of Ps is around \( 6.8 \, eV \), and it can be formed by a reaction between the thermalized positron and one of the excess electrons in the positron track. And it happens in competition with other spur processes, such as excess-electron positive-ion recombination, excess-electron and positron diffusion out of the spur, and excess-electron and positron reactions with the molecules, ions, etc. After thermalization the positron diffuses in the sample for \( 100 - 500 \, ps \) before annihilation. Typically, in condensed matter physics, the energetic positrons injected into condensed matter are slowed down to thermal energy in a very short time of about \( 1 \, ps \). They penetrate into \( 10 - 1000 \, \mu m \) into condensed matter. Thermalization occurs by creation of excess-electron positive-ion pairs along the track. During this process, the last part of this radiation damage along the track will take place at a short distance from the thermalized positron.

There are two forms of positronium depending on the spin of electron-positron pair: one is singlet and the other triplet. When the spins of both electron and positron are anti-parallel to each other, then they form a singlet state \( ^1S_0 \) with total spin 0. That is called para-positronium (p-Ps). The typical lifetime for p-Ps is \( 125 \, ps \). Similarly, when the spins of electron and positron are parallel spin, then they form the triplet state \( ^3S_1 \) with total spin 1. The triplet state is called ortho-positronium (o-Ps). Fig 3.3.1 shows the structure of p-Ps and o-Ps. Typically, o-Ps is more likely than p-Ps). In vacuum, o-Ps annihilates into three \( \gamma \)-rays.

![Fig 3.3.1: Positronium (Ps) is formed depending on the spins of electron and positron and parallel spin Ps is more likely and longer lifetime than anti-parallel Ps.](image-url)
with a mean lifetime of $142 \, n s$. However, in condensed matter, the positron in o-Ps annihilates mainly with an electron of the molecules having opposite spin to that of the positron into two $\gamma$-rays of about $0.511 \, MeV$ each. Then the mean o-Ps lifetime is about $1 - 5 \, ns$. This is called pick-off annihilation. During their lifetimes, the free positron, p-Ps, and o-Ps can react with atoms, ions, and molecules of the sample and/or become trapped in defects in solids. It happens normally changes in the properties of the emitted annihilation photons. Therefore, by measuring the changes the information on the processes can be obtained which have influenced the states of the particles before the annihilation. In addition, the properties of the annihilation photons depend on the states of the particles in the bulk of the sample, because it can be happened that the particle annihilate before the reaction if the concentration of the reactant is very low. Fig 3.3.2 shows a flow chart which describes the map of positron interaction with matter.

Fig 3.3.2: Flow chart of the positron injected into a metal.
3.4 Reference

4 Experimental setup and analysis

4.1 Basic experimental setup

In Fig. 4.1.1, the schematic diagram of the experiment is shown. We use two gamma ray detectors in order to detect two distinct energy gamma rays; one is during creation of positron which is high energy, 1274 keV $\gamma$-photon, and the other is during annihilation of positron that is comparatively low energy, 511 keV $\gamma$-photon. The detectors are set into high voltage and one detector is tuned to the 1274 keV $\gamma$-photon while the other detects the positron’s annihilation radiation (511 keV). The whole experiment is divided into two branches: called fast branch and slow branch. The fast branch processes the time information that measures the time difference between the creation and annihilation of positrons. The slow branch is designed for the energy discrimination of $\gamma$-photons. The effect of slow branch is to tune one detector to the 1274 keV $\gamma$-photon while the other is tuned to the positron’s annihilation radiation (511 keV). The fast branch is comprised of the constant fraction discriminators (CFD) and time-to-amplitude converter (TAC). The TAC records the time between a Start and a Stop signal where the ‘Start’ signal represents the creation of the positron (1274 keV) and the ‘Stop’ signal is for annihilation radiation of positron (511 keV) respectively. The TAC sends the data to the multi-channel analyzer (MCA) that stores the valid events. The slow branch consists of pre-amplifier along with linear amplifier (LA), single channel analyzer (SCA), coincidence unit (COINC) and gate and delay (G & D). The amplifiers are responsible for the amplification and reshaping of the pulses and the SCAs are required for energy discrimination. One SCA is set to 1274 keV $\gamma$-photons and the other is for 511 keV $\gamma$-photons. Both SCAs send the valid signal to COINC unit where it checks the coincidence time of the two events. The COINC unit sends the signal to G&D and the gate opens if it is valid signal and that information reaches to MCA, and thus, MCA is ready to take data from TAC.

We use $^{22}$Na as the positron source for the experiment and the chemical form of $^{22}$Na is NaCl. We bought it from Eckert and Ziegler when it had radioactivity of 30 $\mu$C that is currently decreased to 7 $\mu$C and it is sufficient for our experiment. The NaCl is deposited onto a 7.5 $\mu$m-thick piece of polyimide film (Kapton) and covered by another piece of polyimide of equal thickness (Fig. 4.1.2a). The polyimide film is stable across a wide-range of temperature, from $-269^\circ$C to $400^\circ$C. However, a number of positrons decay within the Kapton film and thus contribute to the overall spectrum. The positron source is kept between the samples under investigation as shown in Fig. 4.1.2b.

![Fig. 4.1.1: Schematic diagram of PALS experiment where HV represents high voltage, PMT for Photomultiplier tube, COINC for coincidence electronics and G&D for gate and delay.](image)

![Fig. 4.1.2: (a) Sealed radiation sources produced. $^{22}$Na sealed between two 7.5 $\mu$m thick Kapton sheets, (b) Sandwich sample setup](image)
4.2 Apparatus description

Detectors:
In Fig. 4.2.1, the gamma ray detectors with the samples and $^{22}$Na radioactive source inside is shown. For the experiment, we use gamma ray detector that is a combination of a scintillator and a photomultiplier tube. It is also called scintillation counter.

The detector is very important and inevitable tool for the experiment. The result of an experiment largely relies on the detector performance. So its proper selection and the corresponding characteristics are necessary tools for the experiment. In nuclear and elementary particle physics, the detectors are defined as a system which absorbs part or all the radiant energy and converts that into some other form more accessible to human perception. The detector performance depends on lots of factors such as sensitivity, response behavior and energy resolution.

Sensitivity:
It is defined as the capability of producing a usable signal for a given type of radiation and energy. None of the detectors are sensitive to all types of radiation at all energies. Typically, the detectors are designed for a certain type of radiation in a given energy range. Within the range, the detectors work very efficiently and outside of it, the efficiency decreases greatly, or they may produce unusable signals.

More generally, the sensitivity of the detector to a given type of radiation and of a given energy depends on several factors pointed out below:
1) the cross-section for ionizing reactions in the detector
2) the detector mass
3) the inherent detector noise
4) the protective material surrounding the sensitive volume of the detectors.

The cross-section and detector mass is very important for ionizing detector like Geiger-Muller counter, because they determine the probability that the incident radiation will convert part or all its energy in the detector into the form of ionization. Basically charged particles are highly ionizing, so even of low density and small volume have some ionization produced in their sensitive volume. However, in case of neutral particles, they first undergo an interaction that produces charged particles which can ionize higher mass density and volume are necessary to ensure a reasonable interaction rate. The noise of the detectors is produced from the detectors and associated electronics. It appears as a fluctuation of voltage or current at the detector output and is always present there independent of radiation.

The material covering the entrance window is very important for the sensitivity of the detector, because the only radiation with sufficient energy can penetrate the layer would be detected. So it relies on absorption coefficient of the material.
Response behavior:
The electrical detector absorbs the radiation and converts that into voltage pulse. It can be operated in continuous mode where the signal is a continuous voltage varying in time with the intensity of the radiation. The relation between the radiation energy and the total charge or pulse height of the output signal is the response of the detector.

The response function of a detector is an important factor for the measurement of energy spectra of which type of radiation being detected. It is the spectrum of pulse heights observed from the detector when it is bombarded by a monoenergetic beam of the given radiation. In our experiment, we use gamma rays which convert into charged particles, i.e.: by photoelectric effect, Compton scattering and pair production in order to be detected. Since the energy of all the photoelectrons is the same, so it results a sharp peak in the pulse height spectrum. However, some gamma rays will also suffer Compton scattering that effects on ideal delta-function. In addition, pair production takes place with those events and changes the total spectrum. Therefore, the observed pulse height distribution will be a combination of the gamma ray spectrum and the detector response.

In addition, the response time of the detector is a crucial factor for the timing properties of the detector. It is the time which the detector takes to form the signal after the arrival of the radiation. The duration of the signal carries similar importance as during that period, a second event cannot be accepted either because the detector is insensitive or because the second signal will pile up on the first. This contributes to the dead time, i.e.: the finite time is required by the detector to process an event which is usually related to the duration of the pulse signal, of the detector and limits the count rate at which it can be operated.

Energy resolution:
It is an important factor which enables the detector to distinguish between two close lying energies. It can be measured by sending a monoenergetic beam of radiation into the detector and observing the resulting spectrum. The ideal spectrum should be a sharp delta peak, however, that comes up with a Gaussian structure with finite width (i.e.: that is expressed by full-width at half maximum, FWHM) due to the fluctuations in the number of ionizations and excitation produced.

Scintillation detectors:
It is one the most widely used particle detection devices in nuclear and particle physics. It makes use of a property of the certain material called scintillator that exhibits scintillation – a property of luminescence when it is excited by ionizing radiation. When an incoming particle strikes the scintillator material, it absorbs its energy and re-emits the absorbed energy in the form of light. If the scintillator is coupled to an amplifying device such as photomultiplier tube, these scintillations can be converted into electrical pulses which can be analyzed and counted electronically to give information concerning the incident radiation.

The basic construction of a scintillation detector is sketched in Fig 4.2.2. Generally, it consists of a scintillation material which is optically coupled to a photomultiplier either directly or via a light guide. When a radiation passes through the scintillator, it excites the atoms and molecules of the material and
makes up the scintillation which causes the light to be emitted. This light is transmitted to the photomultiplier where it is converted into weak current pulse of photoelectrons which is then further amplified by an electron multiplier system. The resulting voltage signal is then analyzed by an electronics system which can give variety of information, such as, sensitivity to energy, fast time response and pulse shape discrimination which are illustrated briefly.

Sensitivity to energy: A scintillator can roughly work as an energy spectrometer. Typically, the light output from the scintillator is directly proportional to the exciting energy which is again directly proportional to the amplitude of the final electrical signal of the photomultiplier. So it can give information about energy of the radiation, although it is not the ideal instrument for this purpose.

Fast time response: The response and recovery times of a scintillator is very short compared with the other detectors such as ionizing detectors. That allows scintillator for precise timing information, such as, time difference between two events. Because of the fast response and recovery time, scintillator detector can accept higher counting rate with reduced dead time.

Pulse shape discrimination: It is a technique of distinguishing between different types of particles by analyzing the shape of the emitted light pulses. Some certain type of scintillator can use this technique by the excitation of different fluorescence mechanism (typically, the fluorescence time is $10^{-8}$ sec; the time roughly taken for atomic transitions, however, if the reemission is delayed because the excited state is metastable, then the process is called phosphorescence or afterglow which can last from a few microseconds to hours depending on the material) by particles of different ionizing power.

The time evolution of the reemission process can be described as an exponential decay,

$$\frac{dN(t)}{dt} = \sum_{i=1}^{m} \frac{N_0}{\tau_i} e^{-\frac{t-t_0}{\tau_i}}$$  \hspace{1cm} (1)

where $N$ is the number of photons emitted at time $t$, $N_0$ the total number of photons emitted, and $\tau_i$ the decay constant that is $i = 1, 2, ..., m$. Since the finite rise time $t_0$ from zero to the maximum in most materials is much shorter than the decay time, so for simplicity, it can be considered as zero. Now if $m = 2$, then the above equation reduces to equation (2) and the corresponding spectrum is given in Fig. 4.2.3,

$$N(t) = A_1 e^{-\frac{t}{\tau_f}} + A_2 e^{-\frac{t}{\tau_s}}$$  \hspace{1cm} (2)

In the above equation the decay constants $\tau_f$ and $\tau_s$ are for the fast and slow components in the spectrum, because most of scintillators show one component is generally much faster than the other. It is also called prompt and delayed components respectively. Their relative amplitudes are given by $A_1$ and $A_2$ which depends on material, however, usually the fast component dominates in the spectrum.
A good detector scintillator is chosen by the following criteria;
1) high efficiency for conversion of exciting energy to fluorescent radiation,
2) transparency to its fluorescent radiation so as to allow transmission of the light,
3) emission in a spectral range consistent with the spectral response of existing photomultipliers,
4) a short decay constant, \( \tau \)

Currently six types of scintillator materials are in use: organic crystals, organic liquids, plastics, inorganic crystals, gases and glasses.

For our experiment, we choose Barium fluoride (BaF\(_2\)) which is presently the fastest known scintillator. It has an emission component with subnanosecond decay time that yields very fast timing, hence good time resolution which is necessary for positron lifetime studies, time of flight measurements, Positron Emission Tomography (PET) and certain high energy or nuclear physics applications. For small detector geometrics, the time resolution can be reached up to 200\(\text{ps}\) by using special electronics\(^1\).

BaF\(_2\) has several scintillation emission bands. The fast scintillation light which is 15% of its light output is emitted in the ultraviolet 195~200\(\text{nm}\) and the decay time of the first component varies between 600 and 800\(\text{ps}\). The slow scintillation component that is the rest of light output centered at 310\(\text{nm}\). The decay time of this component has an average value of 630\(\text{ns}\) (Fig. 4.2.4). The existence of a fast component makes it possible to use such crystals in nuclear physics to measure the time of flight of elementary particles.

In order to detect the fast scintillation light, it is necessary to use a photomultiplier tube with a quartz entrance window. It is necessary to use the optical coupling compound which has good transparency for ultraviolet light. Usually silicon oils or compounds are used for this purpose. The total number of photons emitted from a BaF\(_2\) crystal is about 12 \(\text{per keV}\) of photon energy.

Photomultipliers:
It is an photoemissive device which converts light into a measurable electric current. They are extremely sensitive and, in nuclear and high-energy physics, are most often associated with scintillation detectors. They also find applications in a variety of fields such as astronomy, medical diagnostics including blood tests, medical imaging, motion picture film scanning (telecine), radar jamming, and high-end image scanners known as drum scanners.

The basic construction of a photomultiplier tube (PMT) is shown in Fig.4.2.5. In order to make a radiation detector, the PMT is coupled with a scintillator that produces photons in response to gamma rays deposited in it. A PMT consists of the cathode that is made of photosensitive materials, the dynode string that is the electron multiplier section, and the anode. During operation,
a high voltage is applied across the cathode-dynodes-anode. Incident photons from the scintillator impinge upon the photocathode that emits electrons by photoelectric effect. Due to high voltage, the electron is directed and accelerated towards the first dynode, where upon striking, it transfers some of its energy to the electrons in the dynode that causes the emission of secondary electrons. The secondary electrons are accelerated toward the next dynode where more electrons are released and further accelerated. Thus an electron cascade is created from the dynode string. At the anode, the cascade is collected to give a charge pulse is shaped and serves as a timing pulse. However, the anode pulse is saturated and no longer proportional to the energy of the detected gamma ray. Therefore, a signal is taken from one of the dynodes where the signal is still proportional to the gamma energy to serve as the energy selection pulse.

For our experiment, we use fast time response head-on type Hamamatsu R2059 Photomultiplier tube that has broad spectral response from 160 – 650nm, however, it has high quantum efficiency within 300~500nm. Its maximum spectral response is at 420nm. The diameter of the tube is 51mm and the window of the PMT is made of synthetic silica glass. For the assembly of the PMT we use Hamamatsu H3177 Photomultiplier Tube Assembly in which the PMT assembly window material is quartz.

In Fig. 4.2.6, the complete NIM module with computer system is shown that is connected with the detectors. It contains two CFDs, TAC, two LAs, two SCAs, coincidence unit, delay amplifier and G&D generator.

**Constant fraction discriminator:** The term discriminator is used to define a device which responds only to input signals with a pulse height greater than a certain threshold value. If this condition is satisfied, then the discriminator issues an output signal; if not, no response is made. The threshold can be adjusted on the front panel, and an adjustment of the width of output signal can be made similarly. The most common use of a discriminator is for blocking out low amplitude noise pulses from photomultiplier or other detectors. Triggering the output signal is the most important factor for the discriminator, because the time relation between the arrival of the input pulse and the issuance of the output pulse should be constant. Therefore, a constant fraction discriminator (CFD) is used for timing the pulses. In this method, the logic signal is generated at a constant fraction of the peak height to produce an essentially walk-free timing signal where walk effect is caused by variations in the amplitude or rise-time of the incoming signals (such that if two signals of different amplitude, but with exact coincident are sent into a discriminator with some fixed threshold, then the difference in amplitude will allow them to trigger in different time. This dependency on amplitude causes the logic signal to walk about). The technique by which constant fraction triggering is achieved is shown in Fig.4.2.7. The incoming pulse, $V_a$ is first split into two with one part ($V_d$) delayed by a time, $\tau_d$ equal to the time it takes for the pulse to rise from the constant fraction level to the pulse peak. The other part is inverted and attenuated by a factor $k$ to give a pulse $V_c = -kV_a$. The two are then summed to produce a bipolar pulse, $V_{out}$. The point at which the signals cancel, i.e., the zero-crossing point, is independent of the pulse rise time pulse height. The efficiency of this technique is very high yielding walk as little as $\pm 20\text{ps}$ over an amplitude range of 100 to $1^2$. For our experiment, we use
Canberra 2126 Constant Fraction Discriminator which has 200MHz count rate and walk is < ±30ps (typically for 100:1 range). It has independent controls for threshold, walk, output pulse width, and operating mode, and provides maximum flexibility for use in any timing application. In addition, it is suitable for NIM module as it can operate from standard NIM bin ±12V and ±24V power supplies. If a ±6V power supply is available, it can be configured to use those supplies for most of its power requirements.

**Linear Amplifier:** The Linear amplifier is coupled with preamplifiers. The basic function of preamplifiers is to amplify a weak signal from a detector and to drive it through the cable that connects the preamplifier with the amplifier. At the same time, it should add the least amount of noise as possible. Since the input of the preamplifier is weak, the preamplifier is mounted close to the detector so as to minimize noise. The linear amplifier is used to amplify the signal from the preamplifier and maintain strict proportionality between input and output amplitudes. For spectroscopic application, pulse shaping is an important factor. The pulse, coming from the preamplifier can be characterized as proportional to the energy. The pulse is lasting for a few µs – 100µs, and if a second pulse arrives within this time, then it will add with the tail of the first pulse and its amplitude will be increased, hence the energy information in the second pulse would be distorted. In order to avoid this, the pulse duration is shortened by reshaping the pulse. In our experiment, we use Canberra 2022 Spectroscopy Amplifier. It works nicely for NIM module with better pulse symmetry, minimum sensitivity of output amplitude to variations in detector rise time, and maximum signal to noise ratio. As preamplifier, we use Canberra 2005 Scintillation Preamplifier which is a charge sensitive preamplifier that collects the charge output from the PMT and send that for the pulse shaping linear amplifier. It has faster rise time which is less than 15 ns.

**Single channel analyzer:** The single channel analyzer (SCA) or differential discriminator (DD) is a device working like a discriminator that sorts incoming analog signals according to their amplitudes, that is, it contains a lower level threshold below which signals are blocked. In addition, it has upper level threshold, above which signals are rejected. Thus, only signals which fall between these two levels trigger a response from the SCA. Hence the opening between the upper and lower levels is usually called the window. Therefore, the SCA can be used for energy discrimination. The SCA output is a logical signal, a pulse of 5V amplitude and 0.5µs duration only and only if the input signal is inside the window. For our experiment, we use Canberra 2035A Constant Fraction Timing SCA.

**Coincidence Unit:** The coincidence unit (COINC) is used to determine if two or more logic signals are coincident in time and it generates a logic signal if true and no signal if false. The term coincident means coincident within a time such that the pulses overlap and this time period determines the resolving time of the coincidence and depends on the widths of the signals and the minimum overlap required by electronics. The electronic determination of a coincidence between two pulses is made by a number of ways. Among them, one method is to use a linear transmission gate and the other one is summing up the two input pulses and the summed pulse passes through a discriminator set at a height just below the sum of two logic pulses. In our experiment, we use Ortec 418A Universal Coincidence unit which has adjustable resolving time from 100ns to 2µs.

![Fig. 4.2.7: Technique for constant fraction triggering where rise times of all signals are the same.](image)
Gate and delay: Gate and delay (G&D) generators are triggerable devices which generate variable width gate pulses. It can be triggered by an input logic signal. We use Ortec 416A Gate and Delay Generator which can provide adjustment of the delay, width, polarity, and amplitude of gating pulses. It is also suitable for NIM module and our experiment as it has time stability allows application in systems that require nanosecond time precision.

Time to amplitude converter: The time to amplitude converter (TAC) is a unit that converts a time period between two logic pulses into an output pulse whose height is proportional to the period. A time measurement by the TAC is triggered by a ‘Start’ pulse and halted by a ‘Stop’ signal. This can be done by a simple method of charging a capacitor that is, the TAC begins to charge a capacitor at the arrival of a ‘Start’ signal and to cutoff this charging when the ‘Stop’ appears. The total charge collected is then proportional to the time difference between the ‘Start’ and ‘Stop’ signals. For our experiment, we use Ortec 566 Time-To-Amplitude Converter which can perform nicely within timing range from 10ns to 2ms. It measures the time interval between pulses to its start and stop inputs and generates an analog output pulse proportional to the measured time.

Multichannel Analyzer: The multichannel analyzer (MCA) is a sophisticated device that sorts incoming pulses according to pulse height and keep count of the number each pulse height occurs in its memory. As described above, the MCA is getting valid input pulse to start its process. To prevent wasting processing time on the noise that is always present on the baseline between pulses, the MCA uses a lower level discriminator with its voltage threshold set just above the maximum amplitude of the noise. When a valid pulse from a $\gamma$-photon exceeds the threshold voltage, the subsequent circuitry is triggered to search for the maximum height of the pulse. The data in the histogramming memory breaks up the energy scale of the spectrum into discrete, digital intervals. These intervals are typically called channels. For the experiment, we use the Nucleus Personal Computer Analyzer (PCA - II) card which is a second generation multichannel analyzer card compatible with most of the IBM computer system. It contains a 100MHz Wilkinson analog to digital convertor (ADC), single channel analyzer (SCA), multichannel scaler (MCS), and on board data memory. It transforms the personal computer into a full featured multichannel analyzer along with the standard software. It also utilizes a windowing technique with pull down menus for user friendly operation. For the minimum requirement of the system, it requires RAM with 512k installed memory, a hard disk, a floppy disk drive and a math process in order to increase the speed of the calculations but it is optional, a VGA graphics card and a VGA monitor. A high resolution live display can be seen through the personal computer monitor. In our experiment, we use 8192 channels. The contents of each channel can be displayed on a screen to give a pulse height spectrum $[2-3]$.

4.3 PALS Data and Analysis

The time dependent positron decay spectrum, $D(t)$, is given by,

$$D(t) = \sum_{i=1}^{k+1} A_i e^{-\frac{(t-t_0)}{\tau_i}}$$

(3)

The value of $i$, $i = 1 ..., (k + 1)$ represents the combination of $k$ different types of defects and the lifetime in the bulk. Each of the $(k + 1)$ components is characterized by the individual lifetime $\tau_i$ and amplitude $A_i$ (where $A_i$ are not normalized, but relative intensities, $I_i$ are). In case that no positron traps (defects) are present in the sample, then the spectrum reduces to,

$$D(t) = A e^{-\frac{(t-t_0)}{\tau_B}}$$

, with $\tau_B$ = bulk lifetime
However, the experimentally obtained spectrum differs from equation (3), because of the finite time resolution of the spectrometer. Mathematically the finite time resolution manifests itself by a convolution of the resolution function with eq. (3). In addition, there is always some small number of accidental coincidences that make up a constant background, \((BG)\) which events are uncorrelated to the lifetime. Therefore, the experimental positron decay spectrum has of the form,

\[
N(t) = G(t) \otimes D(t) + BG
\]

where \(\otimes\) represents the convolution operator.

The resolution function of our spectrometer, \(G(t)\), is well-described by a Gaussian,

\[
G(t) = \frac{1}{\sigma \sqrt{\pi}} e^{-\left(\frac{t-t_0}{2\sigma}\right)^2}
\]

The standard deviation, \(\sigma\) is related to the width of \(G(t)\). Experimentally it is convenient to express \(\sigma\) in terms of the full width at half maximum, FWHM,

\[
FWHM = 2\sigma \sqrt{\ln 2}
\]

The convolution of a Gaussian with exponential can be expressed in closed form, and the exponential decay spectrum is given by,

\[
D(t) = \sum_{i=1}^{k+1} \frac{A_i}{2} \exp \left( - \frac{t-t_0 - \frac{\sigma^2}{4\tau_i}}{\tau_i} \right) \left[ 1 - \text{erf} \left( \frac{\sigma}{2\tau_i} - \frac{t-t_0}{\sigma} \right) \right] + BG
\]

where \(\text{erf}(x)\) is the error function.

To obtain values for \(\tau_i, A_i, t_0, \sigma\) and \(BG\), the observed spectrum, \(O(t)\) is fitted to the calculated spectrum \(D(t)\) of equation (6) using a least-squares fit program based on the Levenberg-Marquardt algorithm (LMA). The iteration method of LMA is to find best fit of the spectrum and the goodness of the fit is measured by reduced \(\chi^2\), that is, how well it fits with set of observation. The reduced \(\chi^2\) \((\chi^2_{red})\) is defined as \(\chi^2\) divided by the number of degrees of freedom.

\[
\chi^2_{red} = \frac{1}{v} \sum \frac{(O(t) - D(t))^2}{\sigma^2}
\]

Reduced \(\chi^2_{red}\) distribution

Here, \(v\) is number of degrees of freedom which is given by \(N - n - 1\) where \(N\) is the number of data points, and \(n\) is the number of fitted parameters. The \(\sigma^2\) represents the variance of observation, and \(O(t)\) and \(D(t)\) represent the observed data and calculated values, respectively. A \(\chi^2_{red} \gg 1\) indicates a poor fit to the model. When the \(\chi^2_{red} > 1\), it represents the fit that has not fully captured the data (or that the error variance has been underestimated). A value of \(\chi^2_{red} \approx 1\) means the extent of the match between observations and estimates is in accord with the error variance. And when \(\chi^2_{red} < 1\), then the model is 'over-fitting' the data; either the model is improperly fitting noise, or the error variance has been overestimated.
4.4  Reference


5 Results and discussions

5.1 Experimental procedure

For this work, 99.999% pure aluminum and lead samples were bought from Alfa Aesar. We also worked with shop-grade aluminum (Aluminum 1180) in order to understand the positron annihilation lifetime behavior in that and compare with the pure sample of aluminum. Before the experiment, each time the samples were cleaned up with regular laboratory methanol and dried it up at room temperature.

To anneal the samples, we used a box furnace (Barnstead/Thermolyne – type 47900) that can operate with a programmed temperature profile within the temperature range 100°C–1093°C. Since our temperature domain is within 150°C–670°C, this furnace is suitable for this work. The temperature ranges for annealing were chosen arbitrarily, depending on the melting point of the metals. For example, the melting point of aluminum is 660.32°C, so we started the annealing process from 300°C, and reached until close to the melting temperature.

For pure aluminum, we could reach up to 645°C, whereas we could not reach beyond 630°C for the commercial-grade aluminum as there were a lot of bubble-like structure showing up on the sample as shown in Fig 4.1.2. The pure aluminum did not form any bubble-like structure which allowed us to reach up to 645°C (the samples are shown in Fig 4.1.1).

We arbitrarily chose the duration for the annealing process for our samples. For aluminum, the pure aluminum samples were annealed for 8hrs each time. Similarly, for commercial-grade (or shop-grade) aluminum samples, we took two groups of it with similar size – one group of samples were annealed for 5hrs and the other group was for 8hrs each time. The pure lead samples were annealed for 5hrs each time. The temperature difference between two annealing process was not constant. We chose the temperature gap such as for pure Al, 400°C to 500°C and then 500°C to 550°C depending on the analysis work of the positron lifetime spectrum as there was no abrupt change happening.
We followed a similar procedure for pure lead, which has a melting point of 327.5°C. We started to anneal the sample from 100°C to 320°C, but no bubble-like structure was formed on the sample (shown at Fig 4.1.3).

The positon lifetime spectrum is convoluted with finite time resolution including background. The spectrum contains a source contribution due to the positron source and the source supporting material (Kapton), positron lifetimes in the bulk and in the defects, positronium formation that happens on the surface of the sample\cite{1-2} and the other uncorrelated events form a constant background. In order to subtract the source contribution from our spectrum, we need to calculate the fraction of positrons that annihilate in the source and in the surrounding Kapton foil, $I_{\text{NaCl}}$ and $I_{\text{Kapton}}$, respectively.

The relative intensity of $^{22}\text{Na}$ was calculated by iterative method in this paper\cite{3} which is $I_{\text{NaCl}} = (4.9 \pm 0.4)\%$. The activity of their source was $13 \pm 1 \muCi$ and it was encased by $7\mu m$ Kapton foil. In this paper\cite{4}, it has been stated that positron lifetime due to Kapton foil does not vary with temperature, so the temperature change in laboratory would not have significant effect on the spectrum. The thickness of our Kapton foil is $7.5\mu m$ with activity of $^{22}\text{Na}$ is $7\mu Ci$. In the paper\cite{3}, they replaced $7\mu m$ Kapton foil with $25\mu m$ Kapton foil and did not notice difference in positron lifetime had obtained before. Thus our positron source including Kapton is suitable to collect some values in order to calculate the positron lifetime due to the source.

For subtraction of the source contribution we followed the procedure developed by Djourelov and Mishera\cite{3}. A brief description follows.

From experiments and computer simulations the authors found that the relative intensity of Kapton, $I_{\text{Kapton}}$ can be given by the following empirical relation,

$$I_{\text{Kapton}}(Z) = a + bZ^c$$

which is shown in Fig 4.1.4 where the simulated data were fit presented with a power function with $a = -0.42$, $b = 5.83$ and $c = 0.242$. 

Fig 5.1.3: Pure Pb sample is annealed at 320°C

Fig 5.1.4: The fraction of positron with Z number is annihilated in the Kapton foil supporting the source $^{22}\text{Na}$ (Figure is taken from ref. 3)
In equation (1), it shows that $I_{Kapton}$ does depend on atomic number, $Z$ and for Al, $Z$ is 13,

$$I_{Kapton, Al}(13) = a + b13^c = 10.4\% \quad (2)$$

$$I_{Kapton, Al} = 10.4\%$$

Similarly, for Pb ($Z = 82$),

$$I_{Kapton, Pb} = 16.52\% \quad (3)$$

Therefore, the fraction of positrons that is annihilated in either $^{22}$Na or Kapton for an aluminum sample is given by,

$$I_{Al} = I_{Na} + I_{Kapton} = 4.9\% + 10.4\% \quad (4)$$

For lead we have,

$$I_{Pb} = 21.42\% \quad (5)$$

The values, $I_{Al}$ and $I_{Pb}$, allow us to calculate the source contribution from the observed spectrum. In the observed positron lifetime spectrum, there are three components, $\tau_1$, $\tau_2$ and $\tau_3$ where the first component, $\tau_1$ is the contribution from the bulk given by,

$$\tau_1 = \frac{1}{\lambda_b + k_t} \quad (6)$$

Here, $\lambda_b$ is the positron annihilation rate in the bulk state and $k_t$ is the total transition rate from the bulk state to defect-trapped state (details on this is given in chapter 3, section 3.2).

The second component $\tau_2$ is for defect-trapped state which is given by,

$$\tau_2 = \frac{1}{\lambda_t} \quad (7)$$

where $\lambda_t$ is positron annihilation rate at defect-trapped state. However, in the real observed spectrum, the second component is a combination of defect-trapped state and source contribution. So we calculate the source contribution by equation (5) and (6) from the part of $\tau_2$, and thereafter subtract that values from the part of $\tau_2$ by a fit program that gives us a set of data that is again analyzed by a fit program and gives us the lifetime of defect-trapped state.

The third one $\tau_3$, is for positronium formation. Typically, positronium is formed in polymers, ionic crystals, liquids and gases. There could be gap between the source and the sample, so the positronium forms.
5.2 Pure Al

The positron annihilation data without source contribution was analyzed by a fit program and the range of the lifetimes come out given as follows,

\[ \begin{align*}
\tau_1 & \quad 155 - 167 \text{ps} \quad \text{From the contribution of } \lambda_b \text{ and } k_t \\
\tau_2 & \quad 338 - 383 \text{ps} \quad \text{From the defects} \\
\tau_3 & \quad 1627 - 2448 \text{ps} \quad \text{From positronium formation}
\end{align*} \]

The mean lifetime, \( \tau_m \) for pure Al is given by,

\[ \tau_m = \tau_1 I_1 + \tau_2 I_2 + \tau_3 I_3 \]

The graph of mean lifetime, \( \tau_m \) with temperature is given in Fig. 5.2.1,

In the Fig 5.2.1, the average lifetime decreases with increase in annealing temperature and at 600°C, maximum decrease is observed. It is happening because the fraction of positrons that annihilate in defect sites decreases and more of the positrons annihilate in the bulk state. The annihilation of positrons in the bulk state is followed by equation (6) which says that the lifetime, \( \tau_1 \) depends on annihilation rate, \( \lambda_b \) and total transition rate, \( k_t \) from bulk state to defect-trapped state. For a given sample, \( \lambda_b \) stays the same, however, \( k_t \) may change. Therefore, the more positrons annihilate in the bulk state means that the total transition rate, \( k_t \) decreases. The change in \( k_t \) depends on the nature of the defects. Since during annealing process, the crystal structure is reformed due to migration of atoms, so the number of defects change. After annealing above 600°C (\( T > 600^\circ C \)), the average lifetime increases, which indicates the formation of more defects that trap an increasing number of positrons. Therefore, the optimum annealing temperature for the 8hrs annealing time is just under 600°C.

![Fig. 5.2.1: The mean lifetime \( \tau_m \) was calculated by equation (9) and plotted that versus temperature.](image)

![Fig. 5.2.2: The intensities, \( I_1, I_2 \) and \( I_3 \) of the lifetime \( \tau_1, \tau_2 \) and \( \tau_3 \), respectively are plotted versus annealing](image)
The intensity graph with temperature of the corresponding spectra is given at Fig. 5.2.2. In that graph, after 600°C, there is some change of \( I_1 \) and \( I_2 \), and \( I_3 \). The relative intensity, \( I_2 \) increases when mean lifetime, \( \tau_m \) increases. However, in case of \( I_1 \), it decreases with temperature. Therefore, new defects are forming at \( T > 600°C \). New defects may also have different trapping rate, \( k_t \). If \( k_t \) of the new defects decreases, that leads to an increase in \( \tau_1 \) (equation (6)) and thus to an increase in \( \tau_m \).

It says the same thing as average lifetime that with increase of annealing temperature up to 600°C, the number of positrons annihilate in the bulk state increases, hence the relative intensity, \( I_1 \) increases, consequently, \( I_2 \) decreases.

### 5.3 Shop-grade Al

The shop-grade Al contains various impurities for example, aluminum 1180 has silicon (0.09%), iron (0.09%), vanadium (0.05%), zinc (0.03%), gallium (0.03%), magnesium (0.02%), titanium (0.02%), manganese (0.02%) and copper (0.01%). So there would be a contribution of these elements in the lifetime spectrum.

We used two samples of shop-grade aluminum; sample 1 and sample 2. We annealed sample 1 at 400°C and above and the duration for annealing was 5hrs. So the mean lifetime graph with source correction of sample 1 is given at Fig. 5.3.1.

In the graph, there is no big difference in the mean lifetime, \( \tau_m \) at room temperature and at 400°C. Therefore, until 400°C, there may no be big change happen in the crystal. The most significant change is above 400°C, and qualitatively shows the same pattern as pure aluminum shown in Fig. 5.2.1. The annealing temperature is below 600°C for the duration of 5hrs annealing.

The intensity graph of sample 1 is given at Fig. 5.3.2; it shows that the number of positron annihilates in the bulk state increases with annealing temperature above 400°C, and stays nearly same for both \( I_1 \) and \( I_2 \) below 400°C.

Similarly, for the sample 2 which was annealed at 300°C and above and the duration of annealing process was 8hrs. The corresponding mean lifetime, \( \tau_m \) graph with source correction is given at Fig. 5.3.3. For sample 2, the graph shows that the change is crystal formation starts at \( T = 300°C \) annealing and shows qualitatively same behavior as with sample 1. The intensity graph of sample 2 is shown in Fig. 5.3.4. The graph shows that the maximum number of

![Graph](image_url)
positrons are annihilated at bulk state at temperature below 600°C. Note that sight increase in $I_2$ just above 600°C, which was also seen in the pure Al data (Fig. 5.2.2) and is also somewhat present in the sample 1 data (Fig. 5.3.2).

Fig. 5.3.2: The intensities, $I_1$, $I_2$, and $I_3$ of the lifetime $\tau_1$, $\tau_2$ and $\tau_3$, respectively are plotted versus annealing temperature.

Fig. 5.3.3: The mean lifetime $\tau_m$ was calculated by equation (9) and plotted that versus temperature.
Fig. 5.3.4: The intensities, $I_1$, $I_2$ and $I_3$ of the lifetime $\tau_1$, $\tau_2$ and $\tau_3$, respectively are plotted versus annealing temperature.
5.4 Pure lead

For the pure lead sample we started annealing at temperature 100°C and the duration for annealing was 5hrs. The range of the positron lifetimes for lead are,

\[
\begin{array}{ll}
\tau_1 & 204 - 234\text{ps} \\
\tau_2 & 397 - 411\text{ps} \\
\tau_3 & 1703 - 2168\text{ps}
\end{array}
\]

From the contribution of \( \lambda_b \) and \( k_t \)

From the defects

From positronium formation

Lead has a much higher density than aluminum (see in chapter 2). The mean lifetime graph of lead is given at Fig. 5.4.1.

Unfortunately, the error bars are too large to allow any significant conclusions. Within the error bars there appears to be no significant change happening over the entire temperature range. A combination of longer annealing time, and longer count time to make up for the increased observation of gamma rays in lead may be required in order to get data that shows a significant change for \( T < 300^\circ\text{C} \).
5.5 Reference

6 Conclusions and future work

The main result of this work is that there is an optimal annealing temperature for aluminum just below 600°C. An annealing treatment at 600°C for 8hrs produced the lowest positron mean lifetime, indicating a minimal defect concentration.

Annealing at \( T > 600^\circ C \) results in an increase of the mean positron lifetime due to increased defect formation as the melting point is approached.

Both the shop-grade aluminum and the pure aluminum show this, although it is more pronounced in the latter.

While we obtained reasonable results from the lifetime measurements on the pure and shop-grade aluminum, the results for the lead samples were inconclusive.

The positron lifetime spectra for each measurement were recorded for 30hrs in order to get sufficient count so we can analyze the data with good statistical interpretation with low error bars. While some of the data points have reasonable error bars (3 – 5%) range data points and some of the aluminum data points and all of the lead data have considerable large error bars, so large that it is no longer possible to draw significant conclusions. It will have to be examined if adjustments on the spectrometer and/or hardware modifications can improve this situation.

The shop-grade aluminum sample 2 gives us a hint of change in structure of the sample at 300°C and the sample was annealed for 8hrs. The subsequent temperatures for annealing were selected depending on the data analysis. As future work, this project on the annealing work can be expanded to a closer set of annealing temperatures, e.g. starting at 300˚C then every 25˚C to 625˚C or 650˚C. That will give more data points in the region where the changes happen. The question to be answered is: can we anneal shop-grade aluminum sufficiently well that the two state trapping model can be applied.

In case of lead, it has higher density than aluminum, and that will have an effect on positron annihilation, because more gamma rays will be absorbed in the lead and thus will not contribute to the lifetime spectrum. Therefore longer data accumulation times are required. In this project, we started annealing the sample at 100°C and reached up to 320°C, however we did not notice any big significant change in there. The duration for the annealing process was 8hrs. In addition to that, the obtained data showed larger error bars. So the annealing work on the lead sample can be expanded to include longer annealing time instead of 8hrs. Since lead has such a low melting point, annealing effects in the 200-300°C range are considerable slower, thus longer annealing times need to be used to achieve changes similar to those seen in aluminum.

Lifetime spectra contains a significant fraction of counts that are due to positrons annihilating in the positron source. These counts must be subtracted from the spectrum before data analysis. In this work, we followed the work of Djourelov & Misheva work on source correction\[^1\]. However, there are different approaches, and we have to study these approaches systematically to determine which works best for the setup we have in our lab. For example, we were assuming that the annihilation rate in our \(^{22}\text{Na}\)-source is similar to the source used in the Djourelov &
Misheva paper. This assumption needs to be verified, however, that requires a well-annealed aluminum sample with only a bulk and a small defect contribution.

For both sample, aluminum and lead, an oxide layer is formed when they are annealed in air. In case of aluminum, oxide layer is very thin and resistant to corrosion. Similarly, due to the formation of surface oxide, lead turns into grey color when it is exposed to air. So in future project, it should be determined how much an oxide layer on the aluminum and/or lead affects the positron annihilation spectrum. This could be done by recording a spectrum immediately after annealing, then remove the oxide layer by mechanical grinding and/or chemical etching, then record another spectrum and compare for differences.
6.1 Reference