CHAPTER 5

ENERGY AND TIME RESPONSE OF STILBENE CRYSTALS TO RADIOACTIVE SOURCES AND HIGH-ENERGY PROTON BEAM

5.1 INTRODUCTION

Organic molecular crystals are used in particle identification using pulse shape discrimination technique. We had studied extensively the scintillation property in view of time and energy response of trans-stilbene to fast proton beam and using laboratory sources. All the works described here, have been carried out in Nuclear Science Center, New Delhi, India. By studying the property of stilbene, which is a good pulse shape discriminator ever observed, the results obtained might be generalized to aromatic organic molecular crystals, which have good part in scintillation pulse shape property with exception of materials which have very less pulse shape discrimination properties listed by Winyard et al (1971). Stilbene scintillator has been widely used as a particle identifier to measure neutron energy spectrum against gamma ray background. The fast decay component of the scintillator with varying specific energy loss is the discriminating characteristics for particle identification and its energy. The zero crossing method pulse shape analysis was originally used for identifying the particles. The recent development in timing technology has extended the usefulness of the method to other radiations such as alpha and fission fragments. The improved electronics and consequent high sensitivity allows us to measure the variation of protons and alpha scintillation rise times with energy.
Scintillation response of stilbene has been evaluated for different heavy ionizing particles (Papadopoulos 1999). Theoretical aspects of rise time and its dependence on particle energy of stilbene scintillator have been discussed (Papadopoulos 1997). Stilbene scintillator has been widely used as a particle identifier to measure neutron flux against gamma ray background (Verbitsky et al. 1978). The pulse shape analysis is used to investigate the energy dependence of proton, alpha and other radiation rise times of stilbene scintillator (Harihars et al. 1991 & 1988). The discriminating characteristics are attributed to differences in the fast component of the scintillation decay times of particles with varying energy loss or stopping power dE/dx (Birks 1964). Linearity of scintillation response of proton (0.35-15 MeV) and electron (0.31-0.99 MeV) has been studied (Smith et al. 1968). The alpha-gamma and the neutron-gamma pulse shape discrimination have been carried out using stilbene scintillator (Rousch et al. 1964). The directional anisotropy in scintillation response has been measured for fast neutrons by changing the direction of incident neutrons (Tsukada et al. 1962).

5.2 THEORETICAL PREDICTION BY PAPADOPOULUS (1999 & 1997) AND BIRKS (1964)

There are significant differences in the scintillation process in organic and in inorganic scintillators. In organic scintillators luminescence is associated with conjugated and aromatic organic molecules, which form molecular crystals in which molecules are bound together by van der Waals forces and retain their individual identity, electronic structure and luminescence. The origin of main scintillation emission, i.e., the medium–fast component, is the excitation of the π-electron to excited singlet states. For these components there is no self-absorption of the crystal.

Ionization quenching is another factor that differs between the inorganic and organic scintillators. In organic scintillators the ionization quenching affects the
intensity of fast scintillation component and changes the shape of the scintillation pulse.

The scintillation response $L$ is

$$L = SE'$$  

(5.1)

where $E'$ is the energy of the particle dissipated in the scintillator and $S$ is the absolute scintillation efficiency. The differential form of the equation (5.1) is

$$\frac{dL}{dx} = S \frac{dE'}{dx}$$

where $x$ is the distance traversed by the particle in the scintillator and $E$ is the particle energy.

$$\frac{dL}{dx} = -S \frac{dE}{dx}$$  

(5.2)

In organic scintillators due to quenching effect the above Equation has been modified by Birks (1964) as

$$\frac{dL}{dx} = \frac{S(-dE/dx)}{1 + KB(-dE/dx)}$$  

(5.3)

where $B$ is a constant which depends on ionization and excitation and $K$ the quenching parameter. The rise time of the photon emission, depending on the rate of scintillation response $dL/dt$, can be defined as the time during which $dL/dt$ is increased.

5.2.1 Transit time of particle

For a non-relativistic heavy charged particle having rest mass $M (\gg m_e)$ atomic number $Z$ and velocity $V$, the energy $dE$ transferred as excitation and
ionization along the path \( dx \) to homogeneous medium containing \( n \) electrons per unit volume, is given to a good approximation by

\[
\frac{dE}{dx} = \frac{4\pi e^2 Z^2}{m_0 V^2} n \ln \left( \frac{2m_0 V^2}{I} \right)
\]  

(5.4)

where \( m_0 \) and \( e \) are the rest mass and charge of an electron respectively, and \( I \) is the mean ionization and excitation potentials of the absorber. Here

\[
n = N(A_1 Z_1 + A_2 Z_2)
\]  

(5.5)

where \( N \) is the number of molecules per \( \text{cm}^3 \), \( A_1, A_2 \) are the number of atoms of the constituent elements, \( Z_1, Z_2 \) are the atomic number of the atoms, and

\[
I = N_1 I_1 + N_2 I_2
\]  

(5.6)

where, \( I_1, I_2 \) are the excitation and ionization potentials and \( N_1, N_2 \) the atomic fractions of elements.

The stopping power as a function of particle energy from Equation (5.4) is written as

\[
\frac{dE}{dx} = \frac{2\pi e^2 Z^2}{m_0 E} \left( \frac{MN}{MI} \right) \ln \left( \frac{4m_0 E}{MI} \right)
\]  

(5.7)

The logarithmic term varies slowly with energy and, to a good approximation for energies above 1 MeV, Equation (5.7) can be written as

\[
\frac{-dE}{dx} = C \frac{E}{E}
\]  

(5.8)
where

\[ C = \frac{2ne^4Z^2}{m_0} \frac{m_0}{M} \ln \left( \frac{4m_0}{MI} E \right) \]  

(5.9)

The constant C characterizes both the stopping element and charged particle. We can write

\[ \frac{dE}{dt} = \frac{dE}{dx} \frac{dx}{dt} \]  

(5.10)

This expression, with the aid of Equation (5.8), gives

\[ \frac{dE}{dt} = -\frac{\sqrt{2}}{\sqrt{M}} \frac{C}{\sqrt{E}} \]  

(5.11)

Integrating Equation (5.11) within the bounds from \( E_0 \) (the incident energy of the particle) to E (the particle energy after transit time t), we find

\[ E = \left( \frac{3}{2} E_0^2 - \frac{3C}{\sqrt{2M}} t \right)^{\frac{1}{\sqrt{3}}} \]  

(5.12)

From Equation (5.12) the transit time as a function of energy is

\[ t = \frac{\sqrt{2M}}{3C} \left[ E_0^{\frac{2}{3}} - E^{\frac{2}{3}} \right] \]  

(5.13)

The value E as a function of distance d can be taken approximately by integrating equation (5.8):

\[ E = \sqrt{E_0^2 - 2Cd} \]  

(5.14)

putting E=0 we find range R of the particle:
\[ R = \frac{E_0^2}{2C} \] 

(5.15)

5.2.2 Thick scintillators

When the width \( D \) of the scintillator is \( D \geq R \) of the particle, the transit time can be found from relation (5.13) by taking \( E=0 \):

\[ t_0 = \frac{\sqrt{2}}{3} \frac{\sqrt{M}}{C} E_0^{\frac{3}{2}} \] 

(5.16)

5.2.3 Thin scintillators

When the width \( D \) of the scintillator is very thin \( (D \ll R) \), \( d = D \), \( d \ll R \) and \( E_2 \gg 2Cd \). So we can write

\[ E^2 = \left( E_0^2 - 2Cd \right)^2 = \left( E_0^2 \right)^2 \left( 1 - \frac{2Cd}{E_0^2} \right) \sqrt{\left( E_0^2 \right)^2 \left( 1 - \frac{3}{4} \frac{2Cd}{E_0^2} \right)} \] 

(5.17)

and from Equation (5.13) we take

\[ t_D = \frac{\sqrt{2}}{3} \frac{\sqrt{MD}}{E_0} \] 

(5.18)

5.2.4 Rise time in organic scintillators

In the case of organic scintillators Equation (5.2) is not valid for \( \frac{dL}{dx} \). Due to the quenching effect, the differential light output is given by Birk's (1964) relation, Equation (5.3). From Equation (5.3) and Equation (5.8)

\[ \frac{dL}{dx} = SC \frac{E + KBC}{E} \] 

(5.19)
We can write

\[ \frac{dL}{dt} = \frac{dL}{dx} \frac{dx}{dt} \]  \hspace{1cm} (5.20)

Combining above Equation with Equation (5.19), we get

\[ \frac{dL}{dt} = \frac{\sqrt{2}}{\sqrt{M}} SC \frac{\sqrt{E}}{E + KBC} \]  \hspace{1cm} (5.21)

The value \( dL/dt \) increases up to a maximum for \( E = E_0 \). This value is determined by differentiating Equation (5.21) with respect to \( E \) and putting the result equal to zero.

\[ \frac{E}{C} \frac{dC}{dE} + \frac{KBC}{2E} = \frac{1}{2} \]  \hspace{1cm} (5.22)

Since \( C = \lambda \ln \mu E \), where \( \lambda \) and \( \mu \) are deduced from the comparison of the latter expression with Equation (5.9), we find

\[ E_\sigma = \frac{kB \lambda \ln \mu E_\sigma}{1 - (Z / \ln(\mu E_\sigma))} \]  \hspace{1cm} (5.23)

We may consider that for any organic scintillator and particle, there is a value of particle energy equal to \( E_\sigma \) for which the intensity of the scintillation emission is maximum.

If the incident particle energy is higher than \( E_\sigma \), the scintillation emission has a rise time which is the transit time between the energies \( E_0 \) to \( E_\sigma \) and is given from the relation for \( E = E_\sigma \):
where $E_q > E_o$

If the incident particle energy is lower than $E_o$, Equation 5.24 gives a negative result. It means that there is no rise time and the scintillation rises instantaneously. However, due to surface quenching effect, $dL/dt$ is less than the value to be expected from Equation 5.21 by a factor $\varphi$ with values between 0.5 and 1 (Birks 1964 and Black 1953). The variation of $\varphi$ with $x$ is given from the Birks relation (Birks 1964 and 1951):

$$\varphi = 1 - \frac{1}{2} \left[ \exp \left( -\frac{x}{\alpha_0} \right) - \frac{x}{\alpha_0} E_i \left( \frac{x}{\alpha_0} \right) \right]$$

(5.25)

where $\alpha_0$ is the mean free path of the excitation energy in the scintillator and $E_i(x/\alpha_0)$ is the exponential integral. Here $\varphi = 0$ at $x = 0$ and $\varphi = 1$ for $x > 2\alpha_0$. For stilbene, the value of $\alpha_0$ was found to be 7 $\mu$. So we can consider that, for a distance of $x = 15 \mu$ from the surface, $dL/dt$ is increased and there is a rise time of scintillation given by Equation (5.17) for $D = 15 \mu$.  

The number of molecules per cm$^3$ in stilbene is found to be $N = 3.86 \times 10^{21}$ and according to Equation (5.5), $n = 3.71 \times 10^{23}$ electrons per cm$^3$. From the diagram of $I/Z$ versus $Z$ (Evans 1955) and the atomic numbers of C and H, the values of ionisation potential of C and H are found to be $I_c = 81$ eV and $I_H = 15.7$ eV, respectively, and from Equation (5.6) $I_s = 50.84$ eV. From Equation (5.9) we find for protons and alphas

$$C_p = 2.27 \times 10^{-10} \times \ln(42.8E_o)$$

(5.26)

$$C_\alpha = 36.27 \times 10^{-10} \times \ln(10.7E_o)$$

(5.27)
For *trans*-stilbene KB = 13.7 cm air equivalent/MeV. Using the Bragg-Kleeman rule we find the range per MeV in stilbene.

\[
(KB)_s = 8.21 \times 10^{-4} \times (KB)_{\text{air}}
\]

\[
(KB)_s = 7.03 \times 10^3 \text{ cm/erg}
\] (5.28)

From Equation 5.23, we find for protons and alphas \( E_{\text{op}} = 8.9 \) MeV and \( E_{\alpha} \cong 162.4 \) MeV. Putting the respective \( S = 0.032 \) value in Equation (5.21) we find for protons in stilbene scintillator:

\[
\left( \frac{dt}{dt} \right)_p = 6285 \sqrt{E \ln(43E)} / (E + \ln(43E))
\] (5.29)

where \( E \) is in MeV.

If \( E_0 > E_{\alpha} \), we use the relation 5.24 to compute the rise time of photon emission for protons and alphas for different values of incident particle energy. In case where \( E_0 < E_{\alpha} \), the relations (5.13) and (5.14) and, by a good approximation, (5.18) can be used with \( D = 15 \mu \), if \( R > 15 \mu \). This relation gives for protons,

\[
r_p = \frac{1.1}{\sqrt{E_0}} p^8
\] (5.30)

The data shows that in the region of energies \( E_0 < E_{\alpha} \) the rise time decreases as the incident proton energy increases and in the region \( E_0 > E_{\alpha} \) the rise time increases with energy.

### 5.2.5 Rise time of the photomultiplier current pulse

Assuming that photomultiplier current pulse is of the same form as the intensity of photon emission, it can be written as
where $\tau_r$ and $\tau_d$ are the rise time and decay time of the scintillation and $Q_0$ is the total charge of the photomultiplier during whole scintillation. Differentiating Equation (5.31) with respect to $t$ and putting the result equal to zero, we find the total rise time of the photomultiplier pulse as

$$i(t) = \frac{Q_0}{\tau_d - \tau_r} \left( e^{\frac{t}{\tau_d}} - e^{\frac{t}{\tau_r}} \right)$$  \hspace{1cm} (5.31)$$

Relation 5.33 indicates a strong dependence of the photomultiplier current pulse rise time on the scintillation rise time and, consequently, on the particle energy. In the case of stilbene scintillator, the medium fast component, which gives the main contribution to the scintillation emission, has a decay of $\tau_d = 33$ ns. The rise time measurement of Harihar et al (1991) is in close agreement with the theory of Papadopoulos (1991). Papadopoulos concludes that the rise time of photon emission and the rise time of the anode pulses

(i) decrease as the energy of the particle increases for stilbene scintillator when $E_0 < E_\sigma$  
(ii) increase as the energy of the particle increases for stilbene scintillator when $E_0 > E_\sigma$.

No experimental data are available for energies $E_0 > E_\sigma$ in the case of organic scintillators to verify the theoretical conclusion that the rise time increases with the incident particle energy (Figure 5.1).
The quenching effect in organic and in inorganic crystals is mainly due to two reasons:

1) Conversion of some of their electronic excitation energy into vibration energy and heat.

2) Interaction with other excited molecules or ions.

A particle passing through a scintillator produces excited and ionized molecules along its track. The excited molecules decay by emitting a photon or by quenching. An ionized molecule, by recombination with an electron forms an excited molecule, which decays as previously described. For an excited molecule, the probability of decay by emission of a photon rather than by quenching depends, for a certain molecule, on its environment. When the ionization density increases in the environment of the track, the probability of photon emission decreases. The quenching by process (2) is accentuated and hence the light output is reduced for a more heavily ionizing particle. The energy $E_\sigma$ corresponds to the maximum of $dL/dt$. In the stopping process when the particle energy $E > E_\sigma$ the photon emission increases and when the particle energy $E < E_\sigma$ the photon emission decreases. We may then
assume that the energy $E_0$ is related to the quenching effect and that electron excitation is the main stopping mechanism for $E > E_0$ and that the ionization process is dominated for $E < E_0$.

We know that from Equation (5.11) and (5.21)

$$dL = -S \frac{E}{E + kb\lambda \ln(\mu E)} dE$$

(5.34)

By integrating above Equation between the values $E_0$ and $E_m$ the value of $L$ is found as a function of $E_0$. $E_m$ is defined from the initial condition where $dE/dt$ becomes negative or $\mu E > 1$. The factor $\lambda$, which includes the $A\lambda^2$ term, defines the response dependence on the mass number and the atomic number of the particle. The value of $KB$ appears to have dependence on the type of the ion ($A\lambda^2$) being greater for protons than for heavier ions. Based on the experimental results of Taylor et al (1951) and using Bragg Kleeman rule, the $KB$ for stilbene has been found already. Scintillation response experiments done by Taylor et al (1951) to other particles including proton and theoretical calculation done by Papadopoulos (1999) presents non-linear plot for energy response (Figure 5.2).

Figure 5.2 Scintillation response curve of stilbene for protons (Papadopoulos 1999)
5.3 EXPERIMENTAL

Based on the above theoretical prediction, experiments were proposed and were carried out in Nuclear Science Centre, New Delhi. The crystals grown in Crystal Growth Centre have been cut and polished in Solid-State Physics Laboratory, Delhi. The crystals were highly transparent (Figure 5.3). The crystals have been cut using the inner diameter cutter with crystal boule contained in glass ampoule. The optical planarity of polished face of the crystal has been checked by observing the fringes with the help of yellow monochromatic light. The polishing of crystal was done using alumina 5 micron grade, diamond paste 3 micron, alumina 0.3 micron grade and gamma polish 0.03 micron successively. Part of the crystals from cut ingot was subjected to X-ray rocking analysis using Philips MRD 900 X-ray diffractometer with copper K-alpha having four crystal monochromator of germanium (220) reflections arranged in (n, -n) settings yielding spherical beam of 12 arcsecs width. The crystal, which shows 29 to 32 arcsecs has been made as an element. The elements were of 1.6 cm dia and 3 cm length.

The experiments have been carried out using 14 MV UD Tandem Pelletron at NSC, New Delhi (Figure 5.4), user facility for universities and institutes of India with 25 MeV proton beam. Since every time the change of beam energy is not possible, because of time consumption and difficulty in stabilization and tuning, the energy of the beam has been fixed at 24.8 MeV. The pelletron NMR value for analyzer magnet is 398.817 MT. The thickness of the gold foils in the radiation biology beam line ie., Light Ion Beam Room II (LIBRII) of Nuclear Science Centre, New Delhi is 4 micron. The formula for energy calculations in the beam line is given by,
Figure 5.3 Crystal elements used in the experiment
Figure 5.3 (contd...) Crystal elements used in the experiment
\[ B = K \left( \frac{\sqrt{ME}}{q} \right) \left( 1 + \frac{E}{3725.9} A \right) \]

\[ B = \frac{1438.9 \sqrt{ME}}{R} \sqrt{q^2} \]

\[ q^2 B^2 R^2 = mE \]

\[ E = \frac{2q^2 B^2 r^2}{m} = \left( \frac{3988.2}{797.14} \right)^2 = 25 \text{ MeV} \quad (5.35) \]

Since the output of the proton beam of 25 MeV passes through 4 micron thick gold foil and air thickness of 11 mm, the final beam energy obtained from the pelletron is 24.8 MeV. The diffuser foils of aluminium were made using 99.9% pure aluminium sheets by rolling to different thickness for respective energy by using rolling machine in Nuclear Science Centre target lab. The thickness of foils were calculated using stopping power calculations and ranges of proton in aluminium which are obtained from SRIM 2000 calculations and the plot of energy versus ranges and thickness corresponding to energy diffusion has been identified. The stripped foils of different thickness have been used for the production of protons of different energies. Initially the experiments have been planned for the energy calibration by using SSBD (Canberra make). SSBD output was fed to the amplifier, the ADC and finally to CAMAC and the data acquisition software used is "Freedom". On the other side, amplifier (Ortec 572) was used and the gain of the amplifier was 100 with shaping time of 1 microsec. The slit window was made to collimate the beam from 3.3 cm (LIBRII beam line) to width ranging from 3 mm to 10 mm.

The stilbene scintillator crystal element made from grown boule was mounted on the photomultiplier tube (Philips make). The Bicron organic scintillator grease has been used for coupling the PMT and the crystal. The crystal has been mounted without any air bubble on Philips XP2020 Photomultiplier, which has transit time spread (TTS) of 230 ps. Then the high voltage power supply from Ortec make has been used for biasing the PMT XP2020. This PMT can be biased up to -2.5 kV. The anode output of the PMT was taken and was given to Transient digitizers Tektronix 620A storage oscilloscope and commercial pulse shape discriminator.
2160A. A program called GPIB has been used for recording the pulse spectrum from the digital oscilloscope. Actual plan is to sample out 5,000 spectral pulses and to isolate the different pulse shape for particles and by using software, the pulses will be analyzed and the particle discrimination will be achieved. The dynode output has been taken for energy pulse analysis and given to amplifier via, CAMAC and the corresponding pulses have been processed. The anode output voltage was kept under -2V. By keeping a low PMT gain, the input of the time pick-off circuits is not saturated. The circuits were switched on, 30 hours earlier than the commencement of measurements in order to reduce the electronic drifts and to give time for stabilization of the electronics. In addition, the anode pulse is processed by using a prototype pulse shape discriminator designed by NSC scientists.

5.4 PULSE SHAPE DISCRIMINATION USING COMMERCIAL MODULE 2160A (CANBERRA)

The purpose of pulse shape discrimination is to suppress gamma rays in neutron detection systems utilizing organic scintillators. The method used in PSD module 2160A is basically a zero-crossing circuit.
The 2160A was developed by TU-Munich, Garching. The unit provides optimum pulse shape separation for liquid scintillation counters. However the applications are not limited to n/γ separation; the 2160A can also be used for particle separation with inorganic scintillators, the photos witches, thick SB diodes and proportional counters. The dc coupling allows high statistical count rate without affecting the resolution. The single width module is easy to use, since only anode signal is required from PMT. Figure 5.5 shows the block diagram of the circuit. The anode pulse of the photomultiplier is integrated and differentiated in an amplifier A1 so that the zero-crossing point of the output pulse is determined by the fall time of the input pulse. The shaped signal is then fed to a high gain limiting amplifier A2. The separation of the zero-crossing points corresponding to gammas or neutrons is now greatly enhanced. The overlap of the amplifier signal and the strobe pulse is formed in gate G. If the strobe signal is placed between the crossing points of gamma and neutron pulses, only neutrons cause the output pulse. Here in this 2160A model, the strobe signal is triggered by the output pulse of constant fraction discriminator (CFD), which is delayed and stretched in D.

![Figure 5.5 Block diagram of PSD](image)

5.4.1 Adjustments

To adjust the unit, the inverted output A2 is used (Figure 5.6a). The time relation of the gamma and neutron pulses at the output of the PSD unit (in strobe
position 1) can be monitored with a time to amplitude converter (TAC) (start signal from CFD and stop signal from PSD) and MCA (Figure 5.6b). Since strobe delay is variable, gamma rejection ratio can be adjusted to the onset of the strobe position 2. The onset of the strobe signal appears as a 'sharp needle' in the time spectrum, since the gate has already been opened by a gamma signal. After determining the discrimination point the gate input is switched back to the inverted output A2. The positive output signal is derived, 300 ns after input pulse. The circuit details are given by Sperr et al (1974).

![Diagram showing the relationship between pulses at various points of the PSD circuit: a) Normal operation b) During adjustment](image)

**Figure 5.6** Time relationship between the pulses at various points of the PSD circuit a) Normal operation b) During adjustment

### 5.5 EXPERIMENTAL SETUP

The energy pulse from dynode is processed by Ortec amplifier (572) with coarse gain 100 and shaping time of 1 microsec. The polar pulse was suitably delayed and matched with the time signal and fed to ADC 811. The anode pulse is given to
constant fraction discriminator model (QUAD CFD 454 CANBERRA). The threshold was kept as -0.106 and the width was kept as 20 ns. Careful walk adjustment was done. The output pulse was delayed and was given to TAC as start pulse. The strobe pulse was given to PSD. The anode pulse was given to PSD input. The PSD model used is 2160A CANBERRA make. Careful zero-crossover adjustment along with strobe adjustment was done to get good neutron gamma discrimination for $^{252}$Cf and $^{60}$Co sources before the commencement of the experiment. The output of the PSD was fed to stop of the time to amplitude converter (TAC) EGG /ORTEC 567. The TAC range was kept as 50 ns. The positive TAC output was delayed and matched with the energy signal and the noise level is decreased in the time signal and fed to ADC 811. Using CAMAC CRATE control, the data were collected in list mode as well as singles mode using Freedom data acquisition software. This provides two-dimensional energy Vs time signal plot with Z-axis representing intensity. Further analysis has been done from the collected list mode and singles mode data using Freedom software. The circuit diagram used in the experiment is given below (Figure 5.7).

![Circuit Diagram](image-url)

Figure 5.7 Experimental setup used in the experiment
5.6 CALIBRATION OF PMT AND ELECTRONICS

Non-linearity and poor pulse shape discrimination pertaining to PMT saturation has been observed at \(-1.8 \text{ kV}\). So the PMT has been calibrated with supply voltages varying from \(-1.5\) to \(-2.0 \text{ kV}\). The Philip’s XP2020 is functional up to \(-2.5 \text{ kV}\) and has been checked by varying the supply voltage. For higher voltages, the PMT has more emission in the initial dynodes but later it saturates in the final dynodes. This saturation will lead to loss of energy information of the incident particle. So, the setup was calibrated with different PMT supply voltages using the neutron sources for timing response (Figure 5.8). Then using 25 MeV proton beam, both timing response and energy response were carried out (Figure 5.9, 5.10). It was found that \(-1.6 \text{ kV}\) was an optimum operating voltage. \(-1.6 \text{ kV}\) biasing of PMT gives best energy resolution and highest figure of merit for p-\(\gamma\) discrimination.

Figure 5.8 Timing calibration using 25 MeV proton + \(^{60}\text{Co}\) gamma source
Figure 5.9  Timing calibration using proton beam + $^{60}$Co gamma source

Figure 5.10  Energy calibration using 25 MeV proton beam
The silicon surface barrier detector (SSBD) and the circuit used for initial calibration of proton energy are shown in Figure 5.11. The SSBD was kept before the collimated beam of proton having the energy of 25 MeV. Then the diffuser foil of different thickness has been introduced to the beam and the corresponding energy spectra have been recorded by using the SSBD. The energy of the proton beam has been reduced in steps of 5 MeV. The calibration spectrum has been displayed in the picture (Figure 5.12).

Figure 5.11  Block diagram of SSBD for energy calibration spectrum of protons
Figure 5.12 Energy calibration using SSBD in steps of 5 MeV up to 25 MeV

The response of silicon surface barrier detector is given by graph below (Figure 5.13).

Figure 5.13 Response curve of SSBD to protons
The energy response of SSBD to protons in this energy range is tried with a polynomial fit. The energy response of the detector is quadratic. For higher energies, the response function contains a small quadratic term.

5.8 ENERGY AND TIME RESPONSE STUDIES FOR PROTONS IN STILBENE

Simultaneous energy and time response measurements were carried out using proton beam in the intervals of 5 MeV. The diffuser foils of different thickness have been used before the collimator to reduce the beam energy from 25 MeV to 5 MeV. At high PMT voltage, the pulse saturation has been observed. The width of the peak decreases with increase in energy. But for inorganic scintillators and semiconductor detectors, the width of the time peak increases with increase in energy. The time peaks shift towards lower channel side and the energy peaks shift towards higher channel side. The width of time peak narrows down with increase in energy similar to results of Harihar et al (1991). It was observed that the shifts in PSD peaks take place towards lower channel side with the increase of proton energy.

The two dimensional energy and time spectra (Amplifier Vs TAC) displays the collective information about scintillation response, time resolution and rise time of the scintillator. This two dimensional spectrum shows time along x-axis and amplifier output (pulse height) along y-axis with intensity along z-axis. The characteristics of the scintillator can be accurately known only if the observed pulse height distribution may be satisfactorily related to the scintillation intensity. The two dimensional energy Vs time are shown in Figures 5.14-5.18 for stilbene at -1600 V PMT biasing and in Figures 5.21 - 5.25 for stilbene at -1800 V PMT biasing. The one dimensional intensity Vs energy plot for protons of energy in the steps of 5 MeV upto 25 MeV is shown in Figure 5.19 for -1600 V PMT biasing and in Figure 5.26 for -1800 V PMT biasing. The one dimensional TAC Vs intensity plot for protons of energy in the steps of 5 MeV up to 25 MeV is shown in Figure 5.20 for -1600 V PMT biasing and in
Figure 5.14  Two-dimensional energy Vs TAC spectrum of 5 MeV proton + $^{60}$Co gamma (-1600 V PMT bias)

Figure 5.15  Two-dimensional energy Vs TAC spectrum of 10 MeV proton + $^{60}$Co gamma (-1600 V PMT bias)
Figure 5.16  Two-dimensional energy Vs TAC spectrum of 15 MeV proton + $^{60}$Co gamma (-1600 V PMT bias)

Figure 5.17  Two-dimensional energy Vs TAC spectrum of 20 MeV proton + $^{60}$Co gamma (-1600 V PMT bias)
Figure 5.18 Two-dimensional energy Vs TAC spectrum of 25 MeV proton + $^{60}$Co gamma (-1600 V PMT bias)

Figure 5.19 Intensity Vs Energy spectrum (-1600 V PMT bias)
Figure 5.20 TAC spectrum showing P-γ discrimination (-1600 V PMT bias)

Figure 5.21 Two-dimensional energy Vs TAC spectrum of 5 MeV proton + $^{60}$Co gamma (-1800 V PMT bias)
Figure 5.22 Two-dimensional energy Vs TAC spectrum of 10 MeV proton + $^{60}$Co gamma (-1800 V PMT bias)

Figure 5.23 Two-dimensional energy Vs TAC spectrum of 15 MeV proton + $^{60}$Co gamma (-1800 V PMT bias)
Figure 5.24 Two-dimensional energy Vs TAC spectrum of 20 MeV proton + $^{60}$Co gamma (-1800 V PMT bias)

Figure 5.25 Two-dimensional energy Vs TAC spectrum of 25 MeV proton + $^{60}$Co gamma (-1800 V PMT bias)
Figure 5.26 Intensity Vs Energy spectrum (-1800 V PMT bias)

Figure 5.27 TAC spectrum showing P-γ discrimination (-1800 V PMT bias)
Figure 5.27 for -1800 V PMT biasing. The energy response curve of stilbene with PMT bias voltage -1800 V and PMT bias voltage -1600 V is shown in Figures 5.28 and 5.29. Straggling effects of about 10% produced were ignored using software cutoff 10%. The straggling in energy and time due to the detector and energy of the particle which penetrates the foils are given by

\[ \Delta E = \sqrt{\Delta E_{eb}^2 + \Delta E_{det}^2} \]  \hspace{1cm} (5.36)

\[ \Delta \tau = \sqrt{\Delta \tau_{eb}^2 + \Delta \tau_{det}^2} \]  \hspace{1cm} (5.37)

In stilbene the rise time increases with dE/dr as the energy decreases. The time resolution gets poorer as the energy decreases. Energy response as function of incident particle energy appears to be linear for -1600 V PMT bias. It was non-linear for -1800 V PMT bias with negative quadratic term co-efficient indicating the PMT saturation at high-energy protons. In the time spectrum both the time resolution and peak position are non-linear functions of the incident energy. For the recoil proton (neutron), they can be obtained by interpolation. This illustrates the use of proton rise time spectroscopy in the neutron spectroscopy. Table 5.1 lists the timing spectroscopic results and Table 5.2 lists energy spectroscopic results.

5.8.1 Discussion

Traditionally, the technique of pulse shape analysis using the zero crossing method is used to sort radiations with different rise times, in particular neutrons and gammas. In a scintillator detector system, the rise time of the output pulse from the photomultiplier is governed by the decay time of the radiation in the scintillator. In stilbene the decay time of the fast component depends on the nature of radiation and hence the specific energy loss dE/dr in the detector. The decay times of gammas, neutrons (protons) and alphas have been measured (Bollinger and Thomas 1961). Gammas have the fastest decay time and hence the shortest rise time, followed by neutrons (protons) and alphas. Decay time measurements are lacking for ions heavier
than alphas, in stilbene. Nevertheless, one might extrapolate the increase in rise time with dE/dr for higher atomic numbers. In zero crossing mode of pulse shape analysis, the crossover time depends uniquely on the nature (Winyard et al. 1971) and hence the specific energy loss dE/dr or rise time and the energy of the radiation. In view of the current sub-nano second technology, this dependence of the pulse rise time on dE/dr

<table>
<thead>
<tr>
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<tbody>
<tr>
<td>Protons (-1600V PMT bias)</td>
<td>5</td>
<td>922</td>
<td>2.8</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>861</td>
<td>1.5</td>
</tr>
<tr>
<td></td>
<td>15</td>
<td>822</td>
<td>1.1</td>
</tr>
<tr>
<td>$^{252}$Cf Neutron</td>
<td>20</td>
<td>796</td>
<td>0.88</td>
</tr>
<tr>
<td></td>
<td>25</td>
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<td>0.61</td>
</tr>
<tr>
<td></td>
<td></td>
<td>873</td>
<td>3.19</td>
</tr>
<tr>
<td>Gamma $^{60}$Co</td>
<td></td>
<td>676</td>
<td>1.53</td>
</tr>
<tr>
<td>C(P, nγ) reaction</td>
<td></td>
<td>887</td>
<td>3.22</td>
</tr>
<tr>
<td>Protons (-1800V PMT bias)</td>
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<td>963</td>
<td>3.9</td>
</tr>
<tr>
<td></td>
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<td>2.17</td>
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<tr>
<td>Gamma $^{60}$Co</td>
<td></td>
<td>677</td>
<td>1.39</td>
</tr>
</tbody>
</table>
Table 5.2  Energy spectroscopy- Energy, peak position of protons at -1800 PMT bias voltage and -1600 PMT bias voltage

<table>
<thead>
<tr>
<th>Radiation</th>
<th>Energy [MeV]</th>
<th>Pulse height [channels]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Protons (-1600V PMT bias)</td>
<td>5</td>
<td>63</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>220</td>
</tr>
<tr>
<td></td>
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<td>420</td>
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<td>20</td>
<td>1604</td>
</tr>
<tr>
<td></td>
<td>25</td>
<td>1816</td>
</tr>
</tbody>
</table>

Figure 5.28  Energy response of stilbene (-1600 V PMT bias)
Figure 5.29 Energy response of stilbene (-1800 V PMT bias)

\[ Y = -504.04 + 145.30223 X - 1.89731 X^2 \]

Figure 5.30 Shift in rise time for protons with energy (-1600 V PMT bias)

\[ Y = 988.8 - 15.02 X + 0.26 X^2 \]
may be explored not only to sort different type radiations but to investigate the spectroscopy of a given radiation type as well. Energy calibration for any radiation is readily available, if the relative response is known as a function of energy. Thus neutron (proton) calibration can be done using alphas of known energy. In a rise time spectrum for a given radiation type, higher energy corresponds to lower dE/dr and hence smaller rise time. Hence as the energy increases the rise time shifts to the lower channel side.

Irrespective of PMT voltage, the shift in rise time is towards the lower channels as the energy increases. But the rise time shift is slightly more linear at -1800 PMT bias voltage (Figure 5.31) than at -1600 PMT bias voltage (Figure 5.30). There exists a greater non-linearity at -1800 PMT bias voltage between energy response and time response due to dynode saturation. The energy response curve exhibits Braggs behaviour leading to higher intensity at the end of the particle track. This is similar to

Figure 5.31 Shift in rise time for protons with energy ( -1800 V PMT bias)
the specific luminescence Vs energy curve predicted by Birks (1964) and Papadopoulos (1999). The response functions for energy have been deduced by Birk (1964), Brooks (1979), Rousch et al (1964) and Taylor et al (1951) and they matched their experimental results with non-linear response. Here the non-linear response due to the PMT saturation at -1800 V does not follow the non-linear response trend of the scintillator expressed by the pioneers (Figure 5.29). From the calibration of PMT, the optimized performance of the scintillator has been identified at -1600 V (Figure 5.28). The entire run has been performed at -1600 V and -1800 V PMT. The results are compared. Since it is a collective effect of PMT and scintillator response, very careful attempt is needed to have optimal performance of PMT to isolate its intervention in the scintillator performance. According to Taylor et al (1951), the energy response for scintillator is given by

\[ \frac{L}{MZ^2} = h \left( \frac{E}{MZ^2} \right) \]  

(5.38)

where, \( h \) is a function of \( E/MZ^2 \) and \( L \) the light output. \( Z \) is the charge of the incident particle, \( E \) is the energy of the particle and \( M \) is the mass of the particle. They deduced that the response is non-linear. It is difficult to express the non-linearity in the scintillation light output with energy in experiments due to electronics saturation, spectral sensitivity of PMT and variance in the dynode gain at different stages (Wright 1954). Fowler's (1955) results on energy response are similar to that of Birks (1964) and Papadopoulos (1999 & 1997), which has been comprehended earlier. The energy response function given by Craun et al (1970) is

\[ L(E) = S \left[ dE[1 + kB(\frac{dE}{dx}) + C(\frac{dE}{dx})^2] \right]^{-1} \]  

(5.39)

For electrons Craun et al (1970) have concluded that the response is linear. The true fluorescence intensity is proportional to the total energy dissipated in the crystal over a measured range. There appear to be no quenching processes dependent upon non-
radiative interaction between ionized or excited molecules for, if present, such mechanisms would produce non-linear response. The non-linear response to external electrons must consequently be caused wholly by surface effects such as reflection of primary electrons and escape of secondary electrons and escape of secondary and soft X-rays (or possibly far ultraviolet photons). In addition, overlap of absorption and fluorescence spectra produces considerable variations in scintillation intensity due to a decrease in the efficiency of collection of fluorescence photons as the depth of penetration of the particle decreases. This occurs because of the fluorescence photons emitted by molecules near the surface escaping through the surface instead of being reabsorbed in the crystal with possible collection after subsequent re-emission.

According to Wright (1954) although the response to heavy particles is non-linear, the conditions which exist for these are quite different from those for external electrons. For the same velocity (which altogether with charge mainly determines the rate of energy loss), these penetrate many times further through the crystal than electrons and in any event do not suffer the multiple scattering experienced by electrons, which keeps a large proportion near the surface; it is unlikely therefore that surface effects are appreciable for heavy particles except at low energies.

The linear response to internally produced electrons shows that quenching by non radiative interaction of ionized and excited molecules in the excitation column is not the mechanism responsible for the non-linear response of the organic phosphors to ionising radiations (Wright 1954). In the case of external electrons this is attributed to surface effects, and for heavy particles to bimolecular process connected with the production of temporarily damaged molecules in the excitation column. But the results of our experiments are different and non-linear response has been sorted out using the thorough PMT calibration. In our experiment apart from PMT gain, contribution due to all other factors for non-linear response is negligible.
Figure 5.32  Two-dimensional energy Vs TAC spectrum of C(P,γ) reaction (-1600 V PMT bias)

Figure 5.33 TAC spectrum showing C(P,γ) reaction (-1600 V PMT bias)
Figure 5.34  Energy response of stilbene for C(P,γ) reaction (~1600 V PMT bias)

5.9  CONCLUSION (FOR PROTONS ENERGY AND TIME RESPONSE)

The experimental results follow the rise time shifts towards the lower channel side (i.e., as the particle energy increases the rise time shift decreases) retaining the trend of results of Harihar et al (1991) up to 25 MeV proton. The results partially rule out the possibility of using the theoretical prediction of Papadopoulos (1999 & 1997) in the high-energy side. According to his theoretical prediction the rise time shift increases with the increase in proton energy after 8.9 MeV. But in our case the rise time shifts still maintain its trend by exhibiting the shift towards lower channels, up to 25 MeV. Basic assumption of Papadopoulos (1997) in correlating the transit time to rise time deduced from specific energy loss behaviour does not correspond to rise time shift with proton energy. Specific luminescence will follow the same trend as specific energy loss. Transit time is in the picosecond region whereas the luminescence decay time is in the nanosecond region. For the same reason the
sudden predominance of quenching loss (term $KB$) and the subsequent sudden shooting up of rise time for proton energy above 8.9 MeV is not observed. The photon emission is due to the singlet decay of the molecules and the population of singlet is dependent on $dE/dx$ which may be quadratic or exponential. Similarly the proton energy response is almost linear and the theoretical prediction expresses some non-linearity. In our experiment we have seen that due to PMT saturation the non-linearity arises and it is linear for -1600 PMT voltage. Similar experiments by Taylor (1951) and Fowler et al (1955) show non-linearity in the energy response and it is difficult to compare our experiment (which exhibits linear energy response) with their experiment (which exhibits non-linear energy response). Here the experiments were carried out with thorough calibration of electronics and the PMT voltage, which gives a linear energy response to protons. The high-energy gamma and neutrons are produced in $C(P,\gamma n)$ reaction and the neutron gamma response study has been carried out and good discrimination between the neutrons and gammas has been observed. In the earlier proton response experiments higher energy gamma photons were observed. To see linear response of gamma and neutrons, high intense neutron and gamma background was created by using the above reaction. The resultant spectrum (Figure 5.32) shows that the response is linear. The intensity Vs TAC for $C(P,\gamma n)$ reaction is shown in Figure 5.33. The intensity Vs energy spectrum for $C(P,\gamma n)$ reaction is shown in Figure 5.34. Energy response in thick scintillator is linear as the pulse height over large time constant will be the same without any dependence on stopping power ($dE/dx$). In thin scintillators the $dE/dx$ determines the energy response of traversed particle. But in thick scintillators the entire energy of particle is lost in the scintillator within few picosecond. The energy lost by the particle is converted into luminescence of few nanoseconds in the scintillator. As an outcome the pulse rise times vary with proton energy upto 25 MeV. This variation may be used to estimate neutron energy. The variation of pulse rise time is sensitive to proton energies upto 25 MeV. Rise time spectroscopy in stilbene offers a new means of investigating nuclear phenomena. With improved timing capabilities of available photomultipliers, the zero crossing method has been extended not only to identify a whole range of radiations including heavy
ions, but also in the spectroscopy of these radiations. When radiations such as gammas, neutrons, alphas and fission fragments have different rise times, they can be resolved in the rise time spectrum. For a given type of radiation, the rise time is sensitive to the variation of $dE/dx$ with the energy of the radiation. The time resolution and rise time shift on the energy of the radiation helps in energy and time spectroscopy of the particles for high energies.

5.10 DETECTION CHARACTERISTICS OF VERTICAL BRIDGMAN GROWN STILBENE CRYSTALS FOR GAMMA RAYS USING $^{60}$Co, $^{137}$Cs AND $^{22}$Na GAMMA RAY SOURCES

In the present experimental study the energy response of the grown stilbene crystal for $^{60}$Co and $^{137}$Cs gamma sources was carried out. The time resolution of BGO-stilbene phosphor combine has been studied using $^{22}$Na (511 keV) gamma source.

5.10.1 Experiment (for response of stilbene to gamma)

The VBT grown crystals were cut and polished by using inner diameter cutter with slow rotation. The element 1 cm thick and 1.5 cm dia was used for study. The response of the detector to radio-active gamma sources is investigated. The trans-stilbene crystal was mounted on the PMT of 1" dia model HAMAMATSU photomultiplier R1924 with optical grease without any air bubble for good transmission of scintillation light to photocathode. The PMT is biased through a set of RC filter used to bias upto -1.0 kV. The output from the detector is used to derive the energy and the time information respectively. The time information is derived by processing the signal through timing filter amplifier (ORTEC TFA-863), which filters the fast time components only. Constant fraction discriminator (CFD) (Canberra 2126Q) gives the time at which the input crosses the set threshold and thereby removes the noise from the required signal. The signal is further handled by the gate and delay generator (ORTEC GG8010), so as to get clean trigger.
The energy information is derived by processing the signal through the shaping amplifier (ORTEC 572 - 0.5 μ sec shaping time constant) and analogue to digital converter (ORTEC-811). The relative performance of the sample in terms of timing resolution was measured by recording the output from time to amplitude converter (TAC) started by the Bicron BGO (2.5" hexagon × 2" long) and stopped by the trans - stilbene sample. The timing resolution (full width at half maximum) of the TAC with the 22Na source is 8.5 ns without gate setup.

5.10.2 Results and discussion (for response of stilbene to gamma)

The scintillation response of anthracene and stilbene to low energy protons and X-rays has been investigated. Both anthracene and stilbene show linearity with pulse height energy Vs X-ray energy and non-linearity for low energy protons (Fowler et al 1955). The experimental data and the theoretical values were evaluated for the response of stilbene to D, H2, He and protons of different energy and it was shown that non-linearity exists between the particle energy and scintillation response (Papadopoulos 1999 & 1997). Due to low atomic number of the elements in trans-stilbene it yields good Compton continuum with sharp Compton edge. The scattering of incident photon energy and the momentum gained by the electron is due to Compton scattering. The maximum energy transfer to the electrons (Compton continuum with sharp Compton edge) for Compton scattering is a vital factor in the scintillation materials made of low atomic number. The maximum energy transfer to the Compton electrons by the gamma ray of energy \( E_\gamma \) is given by relation

\[
T_{\text{max}} = \frac{E_\gamma}{1 + 2\alpha} \tag{5.40}
\]

where \( \alpha = \frac{E_\gamma}{m_e c^2} \).
Using the above relation, the maximum energy that can be transferred to electron by means of gamma rays from $^{137}$Cs and $^{60}$Co sources are deduced. By using the gamma ray energies 662 keV, 1173.21 keV and 1332.47 keV, the maximum energy transferred to the electrons ($T_{max}$) was determined by using the above relation and calculated Compton edges are 477.65 keV, 963.2 keV and 1117.62 keV respectively. The energy spectrum (Figure 5.35) obtained using two sources exhibits peaks at 1002, 1094 and 1129 channels respectively. The Compton edge points were taken from the hump peak of the Compton edge from the Freedom software. When the graph is (Figure 5.36) plotted between the channel number and maximum energy transferred (also known as Compton edge), all the three data points were collinear. In Figure 5.36 the X-axis is the calculated Compton edge (from Equation 5.47) (the maximum energy that can be transferred from the gamma energy to electron) and the Y axis is the measured Compton edges in channels from Figure 5.35.

![Figure 5.35 Energy resolution of stilbene for $^{60}$Co and $^{137}$Cs source](image-url)
Figure 5.36 Plot of $T_{\text{max}}$ Vs channels

Figure 5.37 Time resolution spectrum of BGO-stilbene using $^{22}\text{Na}$ gamma source
The time resolution studies were carried out for BGO-stilbene combine using $^{22}\text{Na}$ (511 keV) gamma by means of TAC setup. The start pulse was given by Bicron BGO crystal standard and the stop pulse was given by stilbene crystal. The time resolution spectrum (Figure 5.37) was recorded also by introducing delay time of 15 ns. The shift in the counts for 15 ns delay was measured and time/channel was found. By using the FWHM of the curve obtained by using TAC setup, it was observed that time resolution for BGO-stilbene combine for $^{22}\text{Na}$ 511 keV gamma ray source was 8.5 ns. Decay time is measured by taking the slope of the stopping curve since TAC is stopped by trans-stilbene. The stopping part is only due to stilbene scintillation decay time. The decay time is given by the formula

\[ \tau = \Delta t / [2.3026 \log(N_1) - \log(N_2)] \]  

(5.41)

where \( \Delta t \) is the differential time from the graph and \( N_1 \) is the count number corresponding to the upper end of differential time and \( N_2 \) is the count number corresponding to the lower end of the differential time (Figure 5.37). The decay time is less because the time resolution was done with BGO, which is slow, but stilbene is having very fast decay time. So the combined effect is seen as 8.5 ns and stilbene alone shows the decay time as 5.1 ns. The decay time for the BGO-stilbene combine was calculated from the time resolution spectrum as 5.1 ns.

5.10.3 Conclusion (for response of stilbene to gamma)

The time resolution studies have been carried out for grown stilbene crystal with 1.5 cm dia and 1 cm thick sample. The time resolution observed was 8.5 ns for BGO - stilbene combine using $^{22}\text{Na}$ (511 keV) gamma source. The energy spectrum was taken for $^{137}\text{Cs}$ and $^{60}\text{Co}$ gamma source and it was observed that the three data points of Compton edges corresponding to respective gamma energies and the channel numbers were collinear. The BGO-stilbene combine can be used for evaluating the decay time of scintillator having decay time larger than 5.1 ns.
5.11 IMPROVEMENT OF DETECTION CHARACTERISTICS OF TRANS-STILBENE CRYSTALS BY IMPROVING CRYSTAL PERFECTION

Progressive strengthening of detection characteristics of trans-stilbene have been attempted by improving crystal perfection. A series of timing resolution studies has been carried out for the trans-stilbene crystals grown under different experimental conditions. The results were compared with the previously reported values. Pulse shape discrimination process has been carried out for $^{241}$Am and $^{252}$Cf sources and good discrimination has been obtained for gamma-alpha and gamma-neutron sources from the grown organic phosphor crystal.

The scintillation decay time of trans-stilbene was measured by Leibson for X-rays and Gamma rays and the decay time of 8 ns at room temperature and 5 ns at liquid nitrogen temperature were recorded (Birks 1964). The lowest decay time of 4 ns was reported by Singer (Birks 1964). Harihar et al (1988) have distinctly reported the experimental arrangement for rise time measurement and also the very earliest report by them on rise time given was about 1.6 ns for gamma ray source (Harihar et al 1991 & 1988). In our experiments we obtained rise time of about 1.3 ns and the decay time is found to be 1.6 ns. This improvement in rise time and decay time is accomplished by the improvements made in the crystal growth process by modifying various growth parameters, which are described in chapter 2 and 3. Many reports in the past reveal that the decay times exhibited by the trans-stilbene crystals were between 4 ns to 9 ns (Papadopoulos 1997, Birks 1964). The authors are successful in growing more perfect trans-stilbene crystals thus improving the detection characteristics as manifested by time resolution studies.

5.11.1 Experiment

Timing and discrimination response of the grown crystals to radioactive sources of gamma, neutron, alpha were attempted. The crystals were mounted on
A 1 inch dia Photomultiplier tube (Hamamatsu R1924) with optical grease for light transmission to photocathode of PMT. The PMT is biased through a set of RC filter bias upto \(-0.75 \text{ kV}\). The output of the detector (Crystal + PMT + Base) is used to derive the energy and time information. The timing information is derived by processing the signal through Timing Filter Amplifier (TFA-863-Ortec make), and Constant Fraction Discriminator (CFD TC 454-Tennelec Make). Basically the CFD gives the time at which the input crosses the set threshold and thereby removes the noise from the signal. Then CFD output is gated and delayed by Gate Delay Generator (GG8010-Ortec make) so as to get clean trigger. The signal processed is given to the start of the time to amplitude converter. With a similar processing of output signal, the stop pulse is generated.

The pulse shape discrimination has been done by using the Pulse Shape Discriminator (PSD) setup. The anode output from the PMT was processed by PSD module (Canberra 2160A Module). In yet another way, the same signal is processed by means of CFD and it was gated and delayed and given to start of the TAC and the stop pulse was the PSD output. The zero cross-over technique for the fast and slow component is being used for the discrimination of the particles and gamma rays. The response of crystals to charged particle was non-linear and it has been detailed by Taylor et al (1951) and Fowler et al (1955). Measurements on the response of the scintillator to electrons, deuterons and protons have been investigated (Smith et al 1968, Titus 1970). The energy response of this organic phosphor was non-linear for various particles of different energies. But organic phosphors are helpful to discriminate the charge particles of different masses. The particle discrimination has been carried out for various sources and by various experiments for the past three decades (Plischke et al 1976, Miller 1968, Rousch et al 1964).

The figure of merit is given by (Winyard et al 1971)

\[ M_{a,b} = \frac{T}{t_a + t_b} \] (5.42)
where $T$ is the separation between the time peaks in ns, $t_a$ is the full width at half maximum (FWHM) of gamma zero crossing peak and $t_b$ is the FWHM of the neutron zero crossing peak. The observed figure of merit is 2.37 and 2.53 for gamma and neutron respectively.

5.11.2 Results and discussion

Timing resolution obtained for the crystals grown by conventional Bridgman technique with single walled ampoule was found to be about 9 ns (Arulchakkaravarthi et al 2002f) and the time per channel obtained with the calibration by introducing 20 ns delay was found as $19 \times 10^5$ ns/channel. The timing resolution measured for conventional Bridgman technique grown trans-stilbene crystal with double wall ampoule was 5.78 ns. The time /channel calibration obtained for above measurement was $14.1 \times 10^2$ ns/channel. The selective self-seeding grown crystals show very good timing resolution of 2.23 ns. The time per channel obtained by calibration was $6.8 \times 10^2$ ns/channel. The decay time was calculated by considering the part of the spectra, which was stopped by stilbene phosphor using the formula given by Equation (5.41). The decay times of the phosphor crystal were found to be 5.1 ns (Arulchakkaravarthi et al 2002f), 2.23 ns (Figure 5.38) and 1.6 ns (Figure 5.39) for the crystals grown by conventional method, double walled ampoule and the selective self-seeded method respectively. The low decay time obtained in our crystal is the best report in the literature and this result is essentially due to the improvement in the crystal quality by the newly developed crystal growth technique by the author (Arulchakkaravarthi et al 2002b).

The analysis of the perfection of the grown crystal by time resolution using $^{22}\text{Na}$ 511 keV gamma ray source clearly indicates that the crystals grown by SSVBT show good timing characteristics. The pulse shape discrimination done by using crystals grown through selective self-seeding for $^{252}\text{Cf}$ and $^{241}\text{Am}$ (Figure 5.40 and Figure 5.41) shows better results. The figures of merit given by the crystals are better...
Figure 5.38  Timing resolution spectrum for Na$^{22}$ 511 keV gamma ray using conventional method grown crystal

Figure 5.39  Timing resolution spectrum for $^{22}$Na 511 keV gamma ray using selective self seeded method grown crystal
Figure 5.40 n-\(\gamma\) pulse shape discrimination spectra of \(^{252}\text{Cf}\) source

Figure 5.41 \(\alpha\-\gamma\) pulse shape discrimination spectrum of \(^{241}\text{Am}\) source
when compared with literature values. Low melting point, poor thermal conductivity, melt-solid interface are some of the factors along with substance purity, which affect crucially the quality of organic single crystal. There is a need for large organic molecular crystal with high perfection for scintillation applications. Unsurmountable amount of difficulties arise in seeding and making the crystal free from twinning and multiple twinning. The selective self-seeding method has eliminated all the difficulties and has resulted in crystals with good detection characteristics.

5.11.3 Conclusion (for improvement of crystal quality and scintillation response)

High performance device quality single crystals were obtained from the selective self-seeding technique. The quality assessment made by timing resolution gives the timing resolution of about 2.23 ns, which is an improvement over 7 ns for the crystal grown by conventional method. The decay time calculated from the timing spectrum (1.6 ns) is the lowest decay time for gamma scintillation for this material. The good n-\(\gamma\) and a-\(\gamma\) discrimination has been obtained for \(^{252}\)Cf and \(^{241}\)Am sources.

5.12 EFFECT OF HOLLOW PORES ON THE DETECTION CHARACTERISTICS OF TRANS-STILBENE CRYSTALS GROWN BY VERTICAL BRIDGMAN TECHNIQUE

The grown crystals were studied for their performance in time resolution setup with two anti collinear gamma rays from \(^{22}\)Na sources. The crystals were subjected to optical microscopic and Scanning Electron Microscopic (SEM) studies.

Quality and perfection of crystal play vital role in determining the detection properties (Sangster et al 1956, Birks 1964). The need for large size defect free crystals for detector applications can be achieved by means of adaptation of suitable technique for growth.
5.12.1 Experiment

The grown crystals were cut and polished by using a slow speed inner diameter-cutting machine. The saw marks (app. 75 microns) were removed by lapping operation using different grade of alumina powder followed by chemo-mechanical polish using CCl₄. The polished crystals were mounted on photomultiplier tube (Hamamatsu PMT R1924) of the time resolution setup as shown in Figure 5.42 by using optical grease without any air bubble which will give rise to the attenuation of scintillation light output. PMT is biased through a set of RC filter bias network to bias up to −1.0 kV. The output from the detector is processed by timing filter amplifier (TFA 863 Ortec make) and constant fraction discriminator (CFD) (Quad CFD TC 454-Tenelec make). The signal is further handled by gate and delay generator (GG8010) - Ortec make, so as to get clean trigger. The relative time resolution of stilbene samples are measured by using the time to amplitude converter (TAC), started by the stilbene and stopped by plastic. The data acquisition and analysis was performed by using in-house Freedom software. The full width at half maximum (FWHM) of the timing resolution curve for $^{22}$Na source is 7.8 ns, 5.3 ns and 2.23 ns for the single walled ampoule grown, double walled ampoule grown and SSVBT grown crystals respectively.

Samples from different portions of the crystal starting from the seeding point to the top portion were subjected to optical micrographical studies. The surface of crystals coated with gold was observed using SEM - Stereo Scan 440 in secondary emission mode.

5.12.2 Results and discussion

The time resolution obtained for single walled ampoule grown stilbene crystal is 7.8 ns. Single broad gaussian fits the entire data of the time resolution curve (Figure 5.43). The time per channel calibration obtained using this crystal is 0.232
ns/channel. The crystal surfaces were observed using optical microscope. The grown crystals possess (001) parallel to the growth direction and were confirmed by Enraf Nonius CAD-4 single crystal diffractometer. The big hollow pores were observed perpendicular to (001) plane. The region near walls was having more striations with more interlacing layers (Figure 5.44). The detector element of the crystal is finished in such a way that it has maximum transmittance in the particle incident direction and PMT and negligible transmittance for layer perpendicular to the particle detection direction such that there will be no scintillation light loss. The optical transmission on the layer perpendicular to optical wave-guide of the scintillator is 0.29 %, which neglects any escape of the scintillation light other than in the direction (radiation incident and scintillation light propagation direction) of PMT. Moreover the PMT coupled crystal was covered with reflecting aluminium sheet. Also the transmission was good (more than 60%) for the transmittance of scintillation light to PMT. The pore defects observed using optical microscope are large in size (ie.,15-80 μm) for the single walled ampoule grown crystals.

Time resolution exhibited by the crystals grown by double walled ampoules is 5.3 ns. The time per channel calibration yielded by this crystal is 0.14 ns/channel. The time resolution curve shows 7 gaussian components. FWHM of each gaussian component, centroid and intensity of fitted time resolution curve are shown (Figure 5.45). This crystal also shows negligible transmittance in the direction perpendicular to light transmission to PMT. Aluminium foil covering was done on the crystal with PMT to nullify the escape of scintillation light output. The foil acts as perfect reflecting surface other than in the particle incident direction (scintillation light propagation direction). There is no change in the transmission compared with single walled ampoule grown crystal in the scintillation light wave-guide portion of the crystal. The optical microscopic studies and SEM (Figure 5.46) studies show hollow pores of size ranging from 5 μm to 1 μm. But the hollow pores density is very less compared to single walled grown stilbene. The selective self-seeded ampoule grown crystal exhibits time resolution of 2.23 ns. The time per channel calibration obtained
Figure 5.42 Time resolution setup

Figure 5.43 Time resolution spectrum of the Single walled ampoule
Figure 5.45 Time resolution spectrum of the double walled ampoule grown crystal

Figure 5.47 Time resolution spectrum of SSVBT grown crystal
Figure 5.44  Optical photomicrograph showing hollow pore with striations of Single walled ampoule grown crystal

Figure 5.46  SEM micrograph of double walled ampoule grown crystal

Figure 5.48  SEM micrograph of the SSVBT grown crystal
by using the SSVBT grown crystal is 0.068 ns/channel. Single narrow gaussian (Figure 5.47) exactly fits the data. The optical microscopic studies show very rare defects of hollow pores and scanning electron microscopic studies reveal only the striations on the regions perpendicular to (001) plane (Figure 5.48) of the surface of the as grown crystal. No remarkable changes have been observed in transmission studies when compared to the single walled ampoule grown or double walled ampoule grown samples. The major factors affecting time resolution and line shape in the scintillation counters are the statistical frequency functions and are governed by five separate processes in cascade (a) light production in the scintillator itself (b) light collection of photocathode (c) production of electrons at the photocathode (d) collection of the photoelectrons of the first dynode (e) multiplication of electrons in the photomultiplier structure (Kai Siegbahn 1968).

Except for changing crystals grown by aforesaid methods, electronics of the timing circuits remained the same. So the changes observed are due to the intrinsic property of the crystals. The distribution of photons from scintillation process has a variance, which is due to effects of inhomogeneous luminescence efficiency throughout the crystal (Breitenberger 1955). Fluorescence studies of the materials also do not show any change in emission or excitation bands, but all the crystals grown by different techniques exhibit distinct changes in the detection properties such as energy response and time response. Also the NMR and FTIR confirm that there was no chemical change in the crystals grown by different techniques. So the typical changes in detection characteristics are attributed to the microscopical hollow pore concentrations in the crystal. Very broad gaussian, multiple gaussian and single very narrow gaussian fit exactly the time resolution curve of the crystals grown by single walled, double walled, and SSVBT method grown crystals respectively. The changes in the time resolution curve for different method grown crystals are attributable to the hollow pore concentrations and their size. The broad gaussian fit for the entire time resolution spectrum observed for single walled ampoule grown crystal is due to more concentrated and big size defects. The broad gaussian is the consequence of
equivalent intensity multi time (different decay time) components exhibited by the defects along with the intensity of the real time component (defect free component), which is the property of the crystal. The multi time components with varied intensity giving different gaussian components are attributed to the lower concentration and small size hollow pores of the crystals, which are grown by double walled ampoule. The real time component intensity of double walled ampoule grown crystal is very high and the in between intensity of the defect time components shows the corrugated peak (asymmetric) with multiple split. Also double walled ampoule grown crystal exhibits good time resolution when compared to single walled ampoule grown crystal. The SSVBT grown crystal is devoid of defect component contribution to scintillation and this leads to the single real scintillation lifetime component to be a major contribution to the time resolution curve. This is indicated by perfect fit of single narrow gaussian fit to the time resolution curve exhibited by defect free SSVBT grown crystal. This is confirmed by the optical microscopic and SEM studies. The local thermal fluctuations due to surrounding ambients give rise to super cooling points, yield micropipes, precipitates or inclusions and axial fluctuations result in the edge striations and cracking. These factors are considerably restricted in SSVBT for high performance of the SSVBT crystal.

5.12.3 Conclusion (for effect of hollow pore in stilbene on time resolution)

The surface analysis shows hollow pore defects of large sizes and with high concentration for the single walled ampoule grown crystal. The hollow pore defects are lesser in number and smaller in size for the double walled ampoule grown crystals and they are nearly absent in the SSVBT grown crystals. Time resolution studies were done for the trans-stilbene crystals grown by single walled ampoule, double walled ampoule and SSVB technique. The time resolution spectra show FWHM of 7.8 ns with single broad gaussian fit for single walled ampoule grown crystal. The double walled ampoule grown crystal shows time resolution of 5.3 ns and exhibits multiple gaussian fit. The SSVBT grown crystal exhibits a time resolution of 2.23 ns and single
narrow gaussian exactly fits time resolution curve. The other studies like NMR, FTIR, UV visible transmission, fluorescence studies exhibit no change in all the above three method grown crystals. So the different time resolution exhibited by the trans-stilbene crystal grown by different methods is attributed only to the size and concentration of the hollow pore defects.

5.13 EFFECT OF LOW ANGLE GRAIN BOUNDARIES ON DETECTION CHARACTERISTICS OF TRANS-STILBENE CRYSTALS

Crystals of trans-stilbene were grown by vertical Bridgman technique and tested by X-ray rocking analysis. Low angle grain boundaries were identified for some crystals and they were tested for the performance of scintillation time characteristics. Their performance is compared with grain boundary free crystals. Some of the crystal samples prepared using SSVBT revealed low angle grain boundaries when subjected to X-ray rocking analysis using Philips X'perto X-ray Diffractometer for (200) plane with a glancing angle of 12.148 degrees. The rocking curve with satellite peak before main peak was obtained for low angle grain boundary crystals. This may be due to some unforeseen thermal fluctuations during growth. The broad rocking curve has been obtained for closely aligned crystals grown by conventional method. The grown crystals were cut and polished using fine alumina polish and CCl₄. The fine finished elements of trans-stilbene crystals of 1.5×2.5×1.5 cm³ were mounted on fast Philips PMT XP2020. The PMT was biased through RC filter network up to -1.8 kV. Philips XP2020 PMT has very good timing properties with transit time spread (TTS) of 250 ps. This PMT has better TTS when compared to the PMT H2341 used by Harihar et al (1993), which has TTS of 370 ps. The output is fed directly to 500 MHz digitizer (Tektronix 620A) having 2 GPs/sec with storage facility. The anode pulse width of the fast scintillation and slow components were measured for ⁶⁰Co and ¹³⁷Cs gamma sources and ²³²Cf neutron sources.
5.13.1 Results (effect of low angle grain boundaries on detection characteristics of trans-stilbene crystals)

The anode pulse measured using scintillation light output for $^{60}$Co gamma sources from defect free selective self-seeded grown crystal shows fast component of width of 2.4 ns and the slow component of width 6 ns with respective pulse heights 600 mV and 250 mV. For $^{137}$Cs source, the pulse width obtained for fast component and the slow component were 1.3 ns and 4.2 ns with respective pulse height 600 mV and 125 mV. The anode pulse for the $^{252}$Cf source exhibits 3 ns to 17 ns widths, with pulse height variation from 570 mV to 90 mV. The conventional method grown crystals of stilbene show fast component pulse width of 4.8 ns for $^{60}$Co and slow component width of 17.4 ns with respective pulse heights as 350 mV and 80 mV. The anode pulse for $^{137}$Cs due to the fast component light output shows 4.4 ns width and due to the slow component it was 15.6 ns with pulse height of 440 mV and 125 mV respectively. For neutron source $^{252}$Cf the anode pulse width exhibited by the crystal was between 7 ns to 24 ns with pulse variation from 450 mV to 80 mV. Some SSVBT grown crystals exhibited low angle grain boundaries in the rocking curve, which also exhibits less intense anode pulse with larger width. The anode pulse width using $^{60}$Co gamma rays in low angle grain boundary crystal for fast component was 2.9 ns and for the slow component it was 9 ns with respective pulse heights 500 mV and 130 mV. The anode pulse width measurements of $^{137}$Cs gamma source are 2.0 ns for fast component and 7.6 ns for slow component with pulse heights 460 mV and 120 mV respectively.

5.13.2 Discussion

The gamma interaction in these types of scintillators is with $\pi$-electronic structure and mostly Compton effect type interaction is more pronounced. But neutron interaction takes place by hydrogen or carbon recoiling. So the defect structures due to closely aligned crystals and low angle grain boundaries have severe impact on the
decay characteristics of the grown crystals. Although the forces between molecules in crystals are weak and short range, overlap between the orbital of adjacent molecules is small, there are substantial differences between the electronic spectra, vibration spectra of free molecule and molecular crystal. The disruption due to such normal periodic lattice band structure has more considerable effect on luminescence property (Wright 1987). The different structural defects may be point defect, line defect, planar defect, grain boundaries and structural disorder. Among them grain boundaries give more distorted lattice structure. So this distorted structure with array of defects causes severe effects in the scintillation light output of the crystal and this produces changes in the different scintillation lifetime component.

5.13.3 Conclusion (effect of low angle grain boundaries on detection characteristics of trans-stilbene crystals)

The *trans*-stilbene crystals were grown by vertical Bridgman technique using conventional method and selective self-seeded method. The defect free crystals and low angle grain boundary crystals and high defect density crystals have been identified using X-ray rocking and optical microscope observations. The defect free crystal shows high intense narrow anode pulse. The pulses are broader with less height for low grain boundary crystals. High defect density crystals show still broader and still less intense pulses. The defects in the crystals have severe effect on the scintillation characteristics of the grown crystal.