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The Liquid Scintillator Beta-Alpha Method

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The Beta-Alpha Coincidence Method

The β-α coincidence measurement of concentrations of ^{228}Th hinges on the last few elements of the chain. The beta decay of ^{212}Bi, with an endpoint of 2.2 Mev, is followed by the 8.95 MeV alpha decay of ^{212}Po. The polonium has a half-life of 305 ns, and so if the beta can be detected a subsequent search for the alpha can be conducted over time scales of the order of microseconds with resolutions of the order of nanoseconds. This means that the background from single, un-correlated pulses, such as the other elements of the chain, cosmic rays and room background is reduced by large factors. There is a similar but much slower (164μs) coincidence in the uranium[1] chain which is essentially the above system with two neutrons added to each nucleus. The possibility of measuring the thorium and uranium chains simultaneously is thus obtained.

The method used by the authors is a detection system based on a liquid scintillator source coupled to a photomultiplier. This has the advantages of experimental simplicity and a necessarily fast recovery time from the initial β pulse. The use of water miscible liquid scintillator allows samples from most water systems to be mixed directly with the detection medium, and maximal geometric efficiency is guaranteed. The absence of any chemical processes in preparation is also a consideration when comparing β–α methods.

The main limitation of such a system is that the maximum efficiency is determined by the branching ratio for the initial β decay. In the case of the thorium chain this is 64%, while for the uranium chain the branch is 99.9%. Counting efficiencies of around 80% have been obtained, with the other 20% being lost to the time cut on the

[1] In the following document 'thorium chain' applies to ^{228}Th or ^{232}Th, while 'uranium chain' applies to ^{238}U or more specifically ^{226}Ra.

164μ / 305n = 5..

10 l H2O
30 ml scint.

radon in scintillator
thicker jars
vacuum N2 flush

30 ml → 2 liter eventually
jar
factor 50
↓
bkg goes up
cosmic goes up

exponential and the low energy betas. Such a number gives efficiencies for thorium counting of around 50%. This efficiency is almost independent of the scintillator used, as the only dependance on light yield is at the low energy beta cut-off imposed by the low level discriminator. The tube voltage can also be adjusted with a fair degree of impunity to increase the beta signal and circumvent this problem. At present two low background channels of a modular system have been assembled, with a further four in production. A drawing of this system is attached. A separate system, consisting of a non-shielded 3" PMT, is being kept for high count rate work.

An anti-coincidence stage has also been added in order to veto the system with respect to cosmic rays. This is necessary as the large number of photoelectrons (of order 2000) produced by a minimum ionizing potential particle travelling through the scintillator is enough to make the discriminator re-fire on the falling edge, and bogus coincidences are seen. Pulse height cuts can also be applied to achieve the same effect, although a hardware option is always preferable.

Source preparation involves adding the aqueous solution to be counted directly to the scintillator and sealing the source. The water/scintillator combination forms a clear mixture after around 30 seconds of shaking and the source is then placed directly on the tube after having been smeared with an optical coupling compound. The tube voltage is applied and counting can begin immediately. The total time involved in preparation can be less than five minutes, so allowing measurements of 10 hour lead. The standard sample holder is a 60 ml PMP jar from Nalgene, the bottom of which is machined to a flat. Using high capacity scintillators, this allows 10 ml of an aqueous sample to be counted. Acceptance tests have been carried out and sample loadings of 25% (ie 15 ml of a total volume of 60 ml) have been confirmed for: 0.1M H_2SO_4, 10% $MgCl_2$ and 1M HNO_3.

Recent work has been on two major areas: the first is the counting of the uranium chain, the second is pulse shape discrimination (PSD). The uranium chain measurement is simple in principal in that the coincidence gate just has to be widened from $1\mu s$ to $500\mu s$. Tests where this has been done[2] show that efficiencies for 2 Bq spikes are around 80%. This corresponds with the 80% counting efficiency mentioned above for thorium and the 99% branching ratio.

Pulse Shape Discrimination

The work on pulse shape discrimination has looked at the systematic differences

[2]See figures attached.

between the alpha and beta pulses due to their respective energy deposition methods. The digitizing of the pulses is at present done with fast digital oscilloscopes, some of which can resolve down to the half nanosecond level. As an order of magnitude, the rise time of the 2 inch tube is around 7 nanoseconds.

Algorithms for separating alphas and betas have been tested by taking true beta-alpha coincidences from spiked sources and looking at the tails of the pulses. The alpha pulse has a much larger tail than the beta, and this difference extends over a fairly large timescale with respect to the pulse width, as shown in the attached figures. Normalisation of this tail area is needed as we have an extended beta spectrum, and the first quantity tried was the peak voltage. Although the separation given by this method was very good, recent work has involved normalisation by the total peak area, and this seems slightly better as the error on the peak area is smaller. This opens up the possibility of using two gated ADC's to measure the tail charge and the total charge, and hence full digitization of the pulse would be unnecessary. The figures attached show average beta and alpha pulses and scatter plots of the tail area against peak height.

As can be seen, a good degree of separation is obvious, even more so in the third figure, which shows the distribution of the given ratio for alphas and betas. Such a graph shows a cross-over of only around ten events in 2000, leading to a pulse shape discrimination of approximately 0.5%. In practice, any events above a certain value would be classed as alphas, while any below another value would be said to be betas, hence leading to almost total separation with a small loss in counting efficiency.

The subject of de-gassing the scintillator has been looked at also, with runs as above for both a scintillator as from the manufacturers and a sample which was purged with nitrogen for a few minutes. Although the peaks shifted slightly, there was little change in the separation, with both samples giving around 0.5% cross over. The extent to which the cocktail direct from the suppliers is gassed or degassed is not known, but the indications are that degassing looks to be unnecessary.

Background

To finish, the subject of the background in the system is reviewed. There are two main areas of background in such a system: the random γ-γ coincidences, and the contaminatory background. The first is related to the room background rate in that non-time correlated singles can occur within 1 μs of each other. In such cases, the rate of random coincidences is related to the square of the singles rate, and hence work has been progressing on the shielding of the source. As can be seen in the figures supplied, a new shielding system has reduced the singles rate to around 0.5 Hz, giving a random rate of 0.025 counts per day in the thorium chain. In the uranium chain, we have to multiply by 500, due to the change in coincidence window, giving a random rate of 12.5 per day. This rate has been confirmed in work not shown here.

We can further reduce the background using energy cuts on the stop pulse, as shown in the figures. Given a factor of around 3 reduction due to the energy cut, PSD discrimination of only around 10% is needed to reduce the random rate to less than one half per day. Given the previous determination of a PSD rejection of $\leq 1\%$, it is clearly seen that γ-γ randoms are no longer a problem.

Contamination backgrounds are different in the sense that a PMT counter cannot be contaminated (in the sense that it has to be cleaned) because sealed sources are placed on the phototubes. Thus the 'contamination' is of the jars themselves. At present, levels of around 3 per day are seen for the thorium chain, and if all of this is assigned to thorium in the scintillator, the contamination is at the 10^{-10} g/g level. This is seen as much to high, and possible causes include such effects as the detergents in the scintillator being preferentially contaminated, insufficient cleaning of the jars, negligence in source preparation and so on. Although these events seem to be real, in the sense that the stop pulses pass energy and PSD cuts, time correlated singles cannot be positively ruled out until larger data sets are available.

Build 6 in early January.

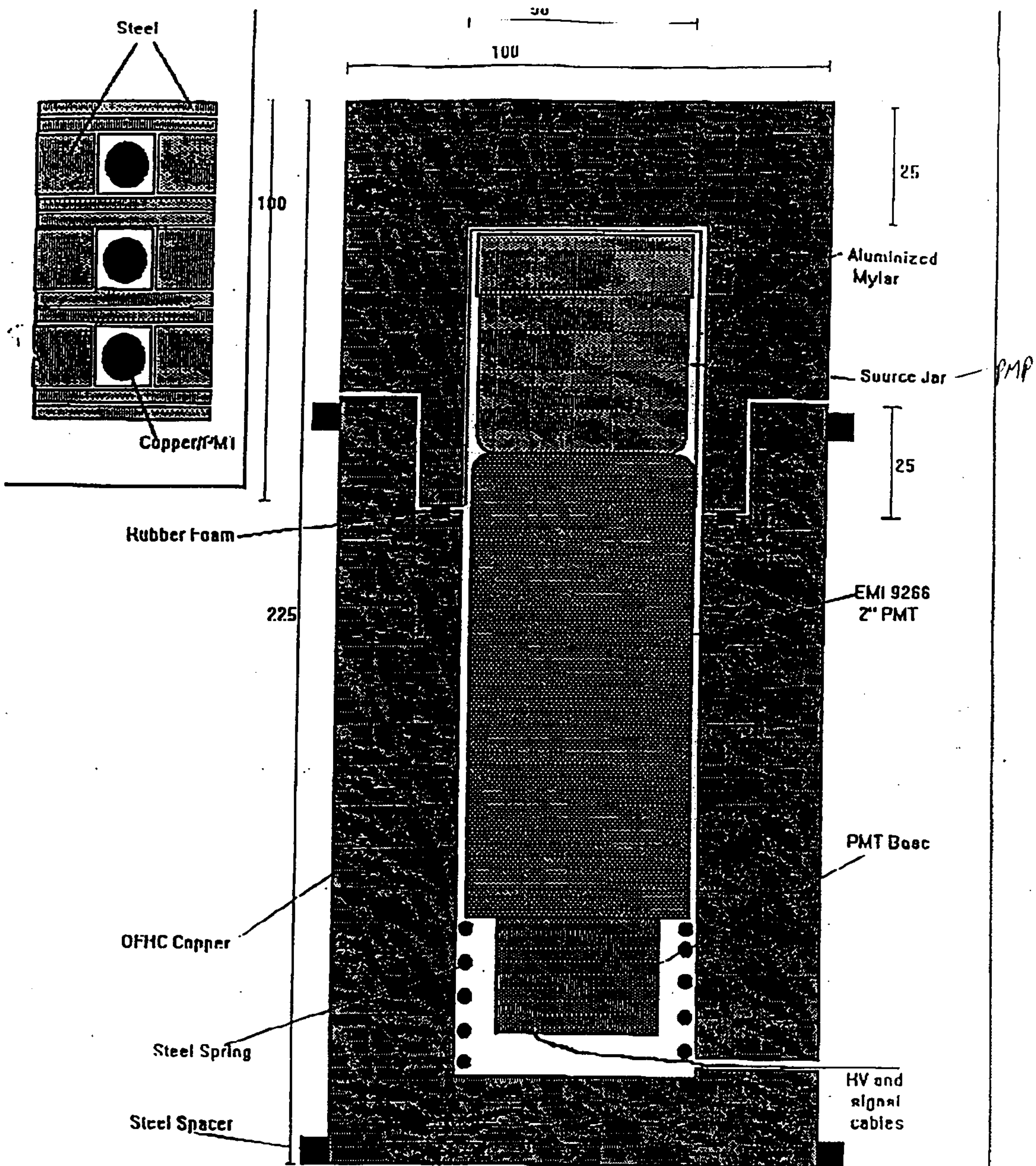
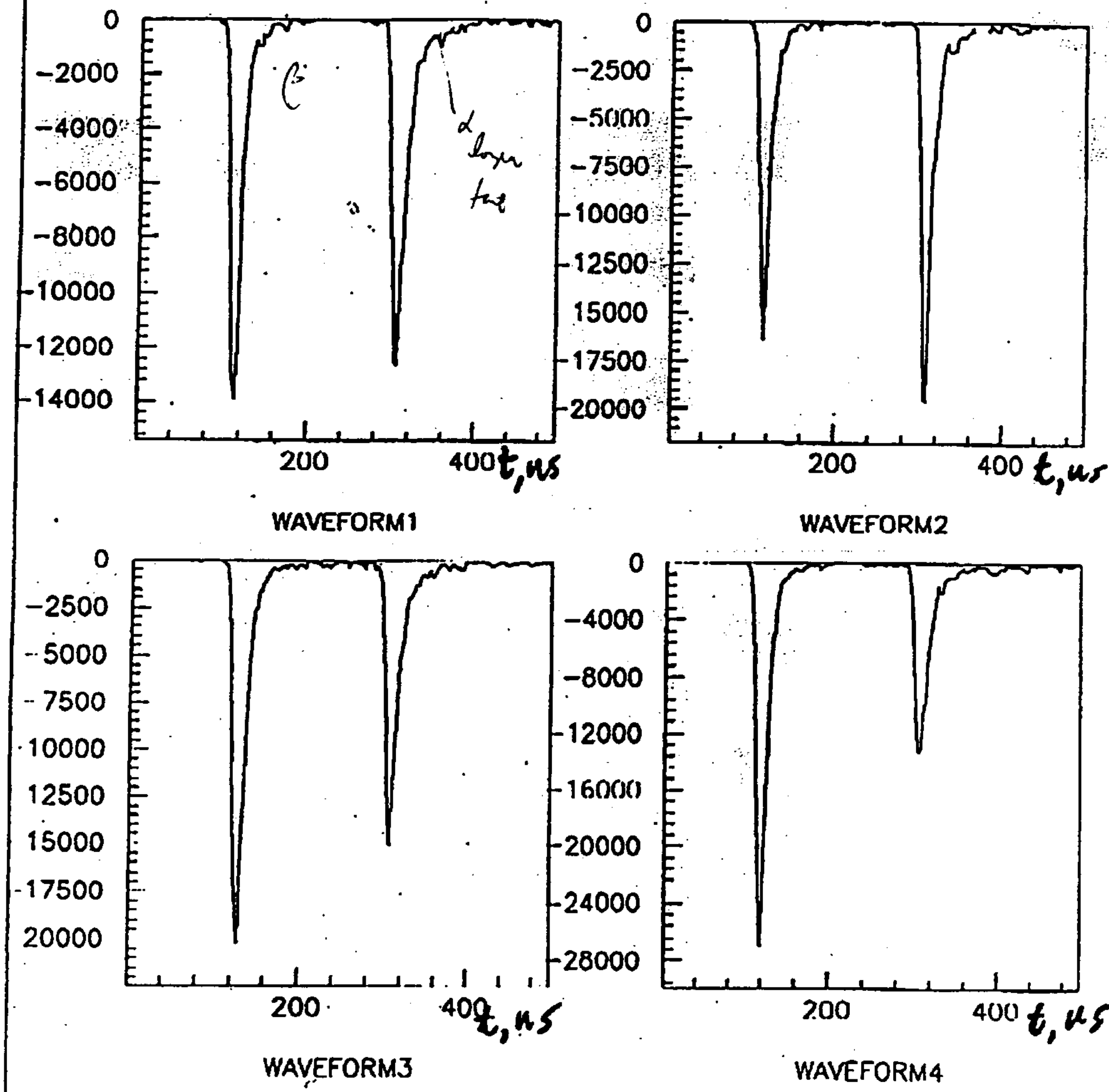


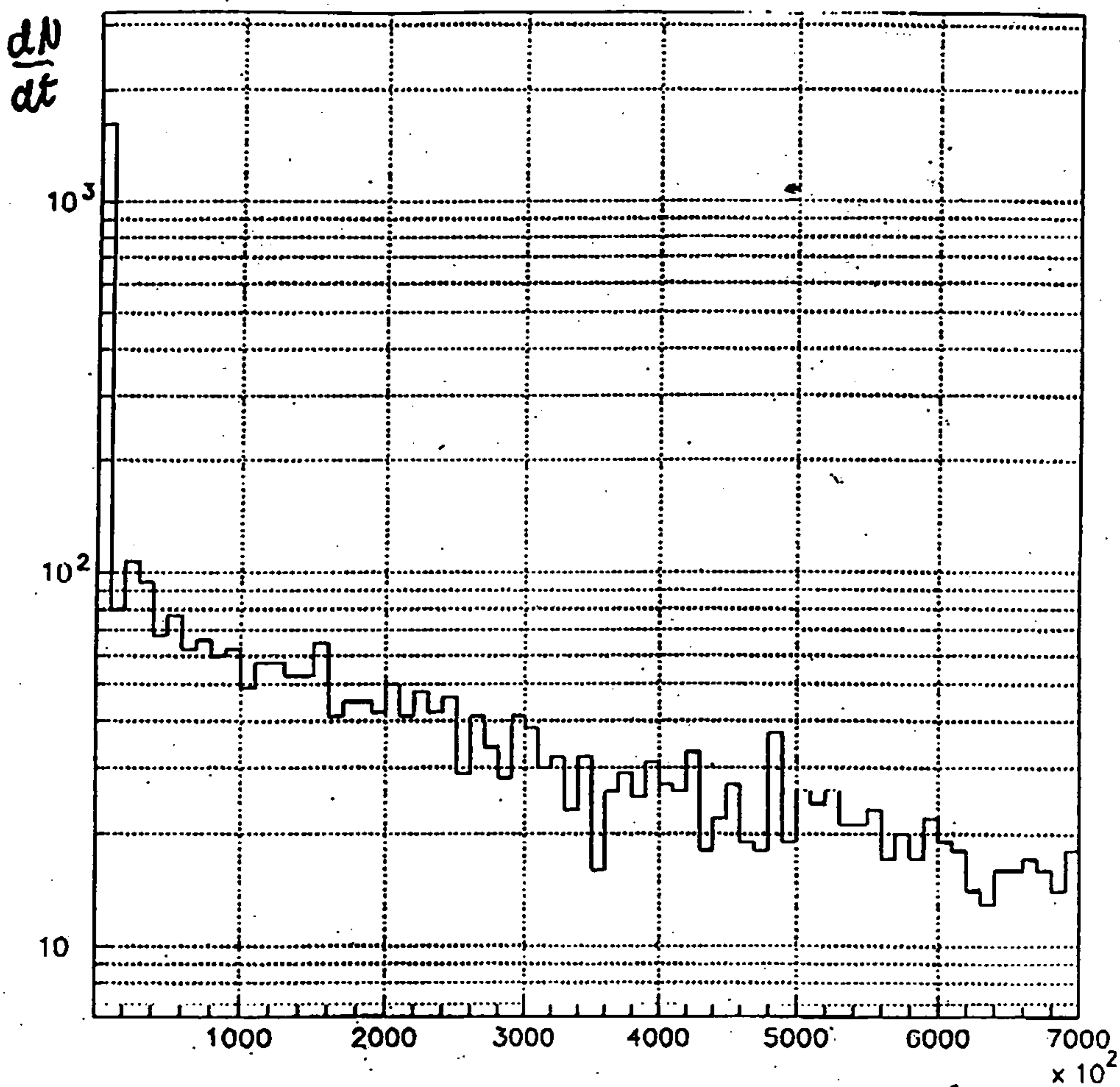
DIAGRAM OF 2" MODULAR PIECE
ALL MEASUREMENTS IN mm
SPACERS ARE USED TO ALLOW CYLINDER TO BE MATED WITH 4½" STEEL BLOCKS AS SHOWN.

Four sample digitised waveforms from sample of 2000. Sample rate 1Gsa/s. Y scale (voltage) is 10 bit enhancement of 8 bit ADC data.

Coincidences were selected to be >200us apart ~~and energy cuts made so that β peak resemble~~ α peak.



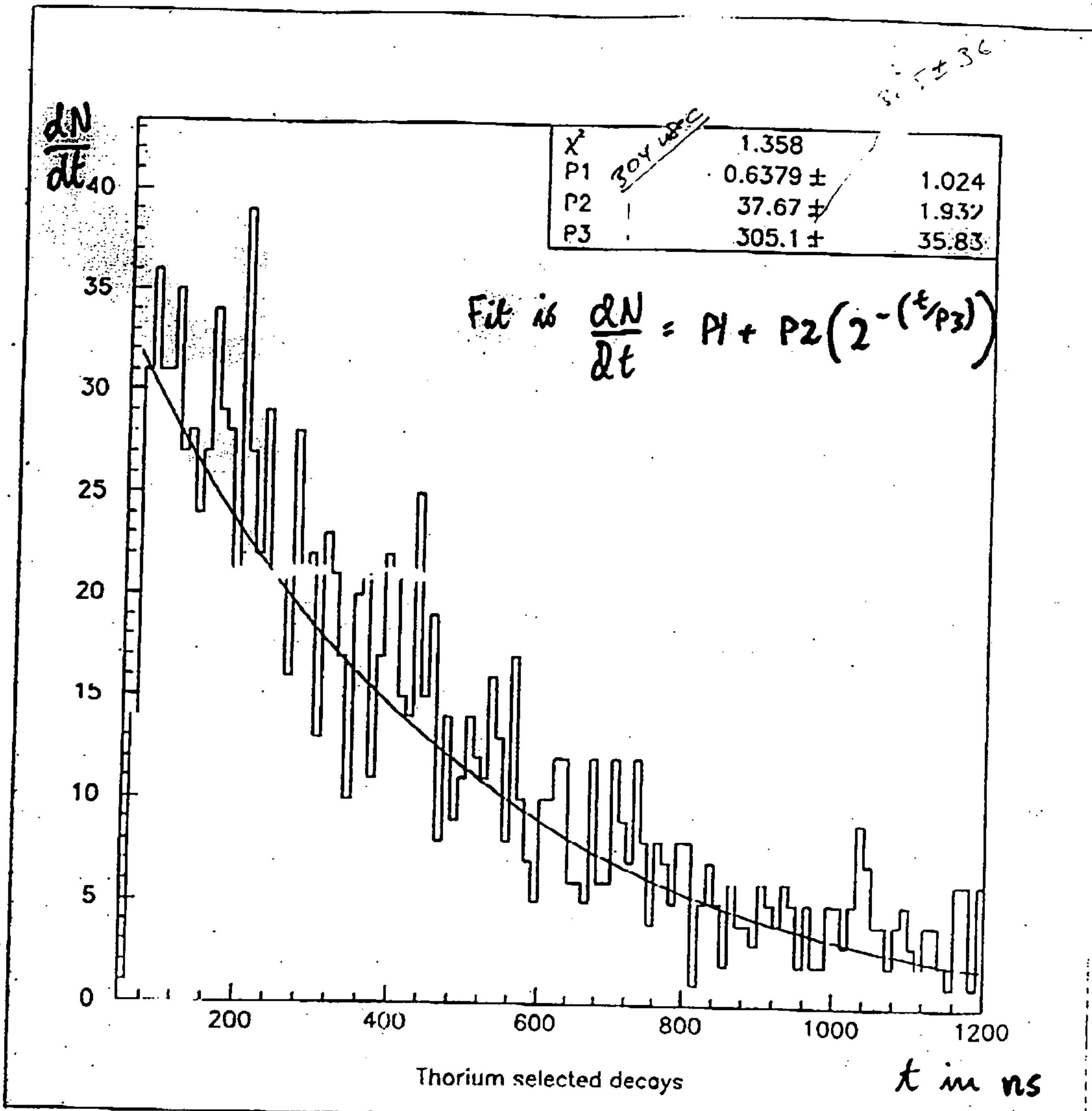
DELAY TIMES between Start & Stop pulses for Spike of 2Bq each ^{226}Ra & ^{228}Th.



Spike of 2Bq each Th and Ra

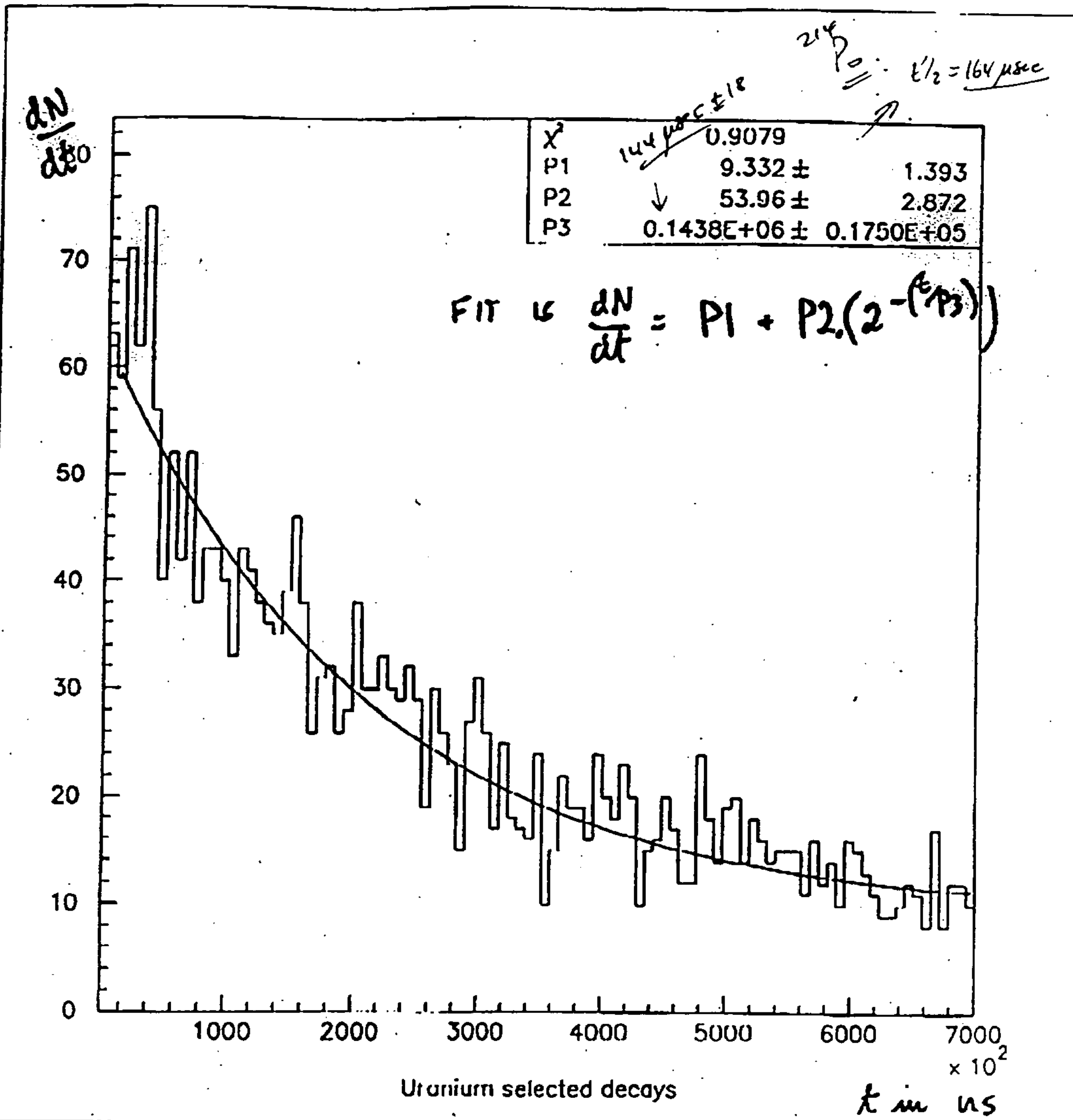
DECAY TIMES SELECTED FOR

t < 1200ns ie Thorium Chain



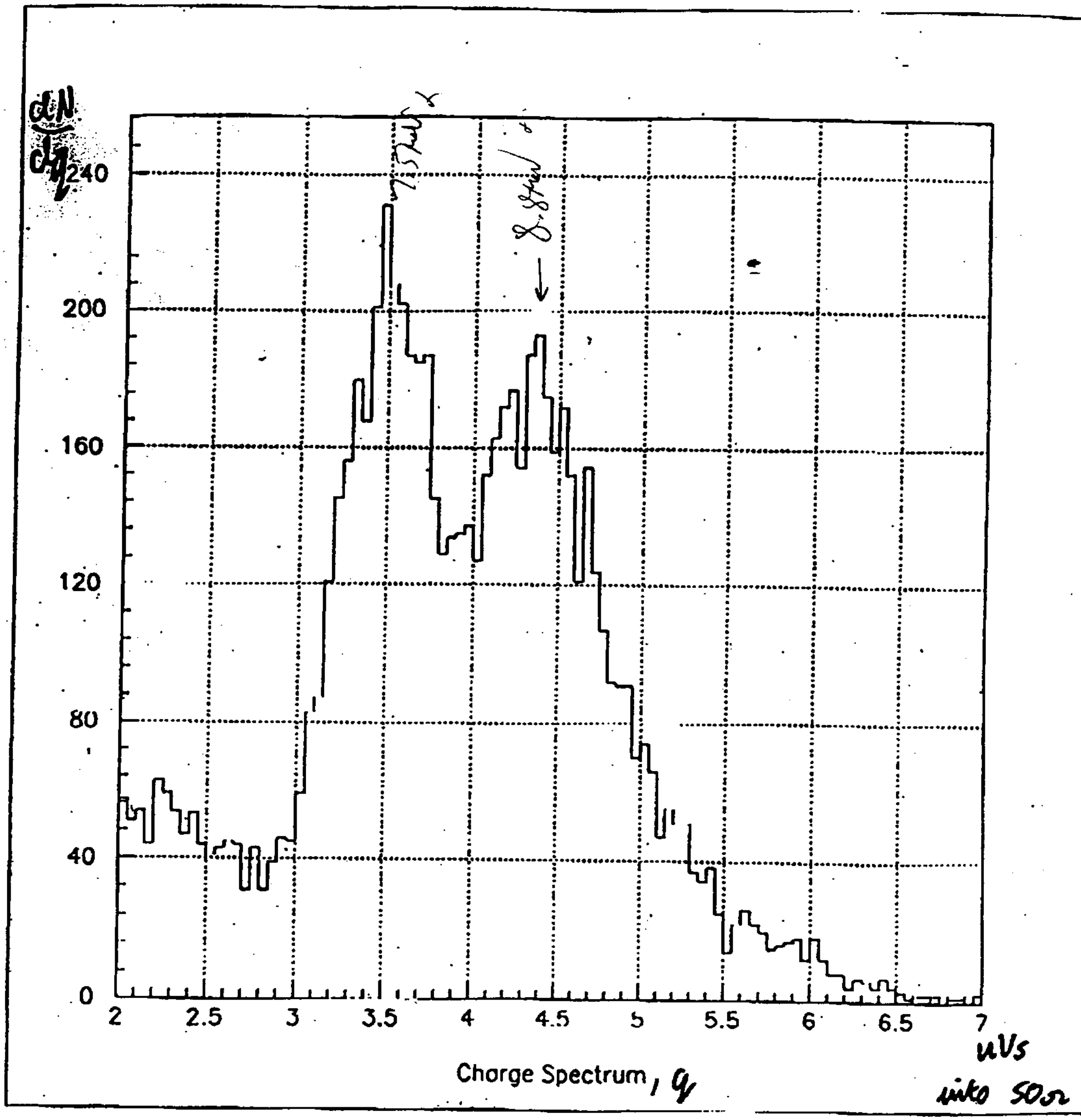
DECAY TIMES SELECTED FOR [redacted]

5μs < t < 700μs ie Uranium chain



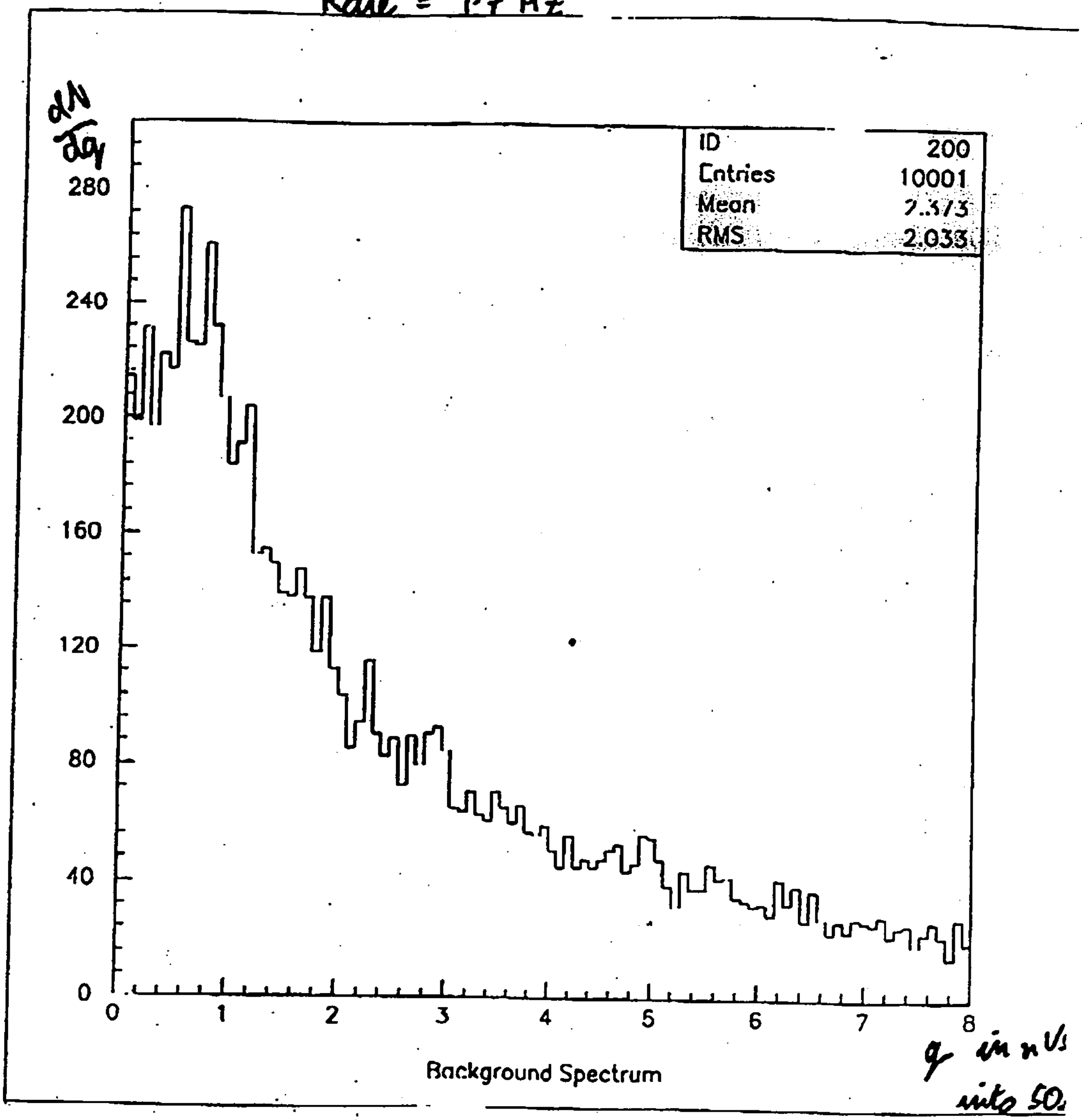
As time windows are the same, the ratio of $P2_{Th}$ & $P2_U$ should be equivalent to the ratio of branching ratios : $\frac{P2_{Th}}{P2_U}$ = 69% cf 6

Charge Spectrum of Stop Pulses for Mixed spike.

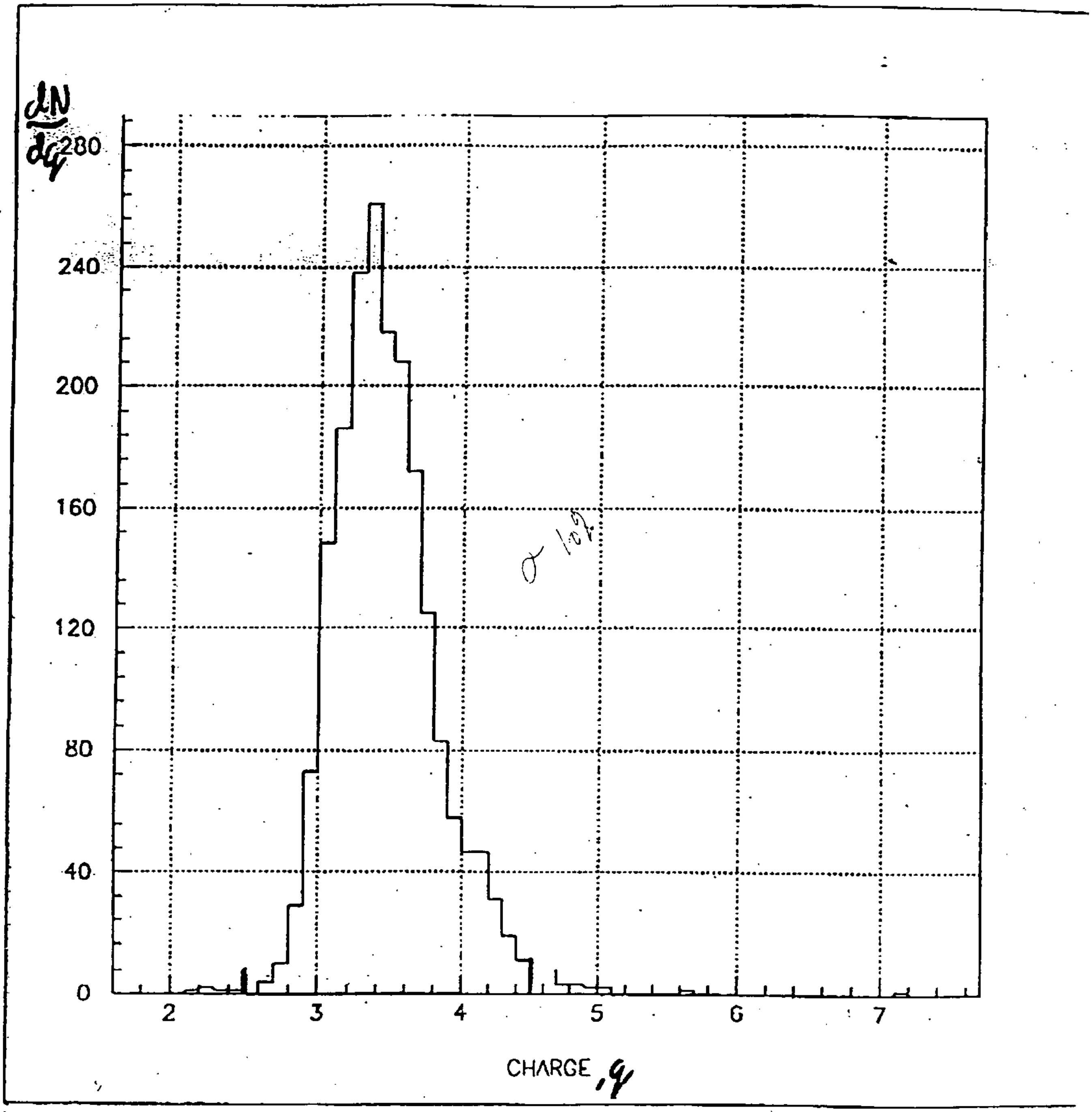


γ Background Charge Spectrum
with copper shielding

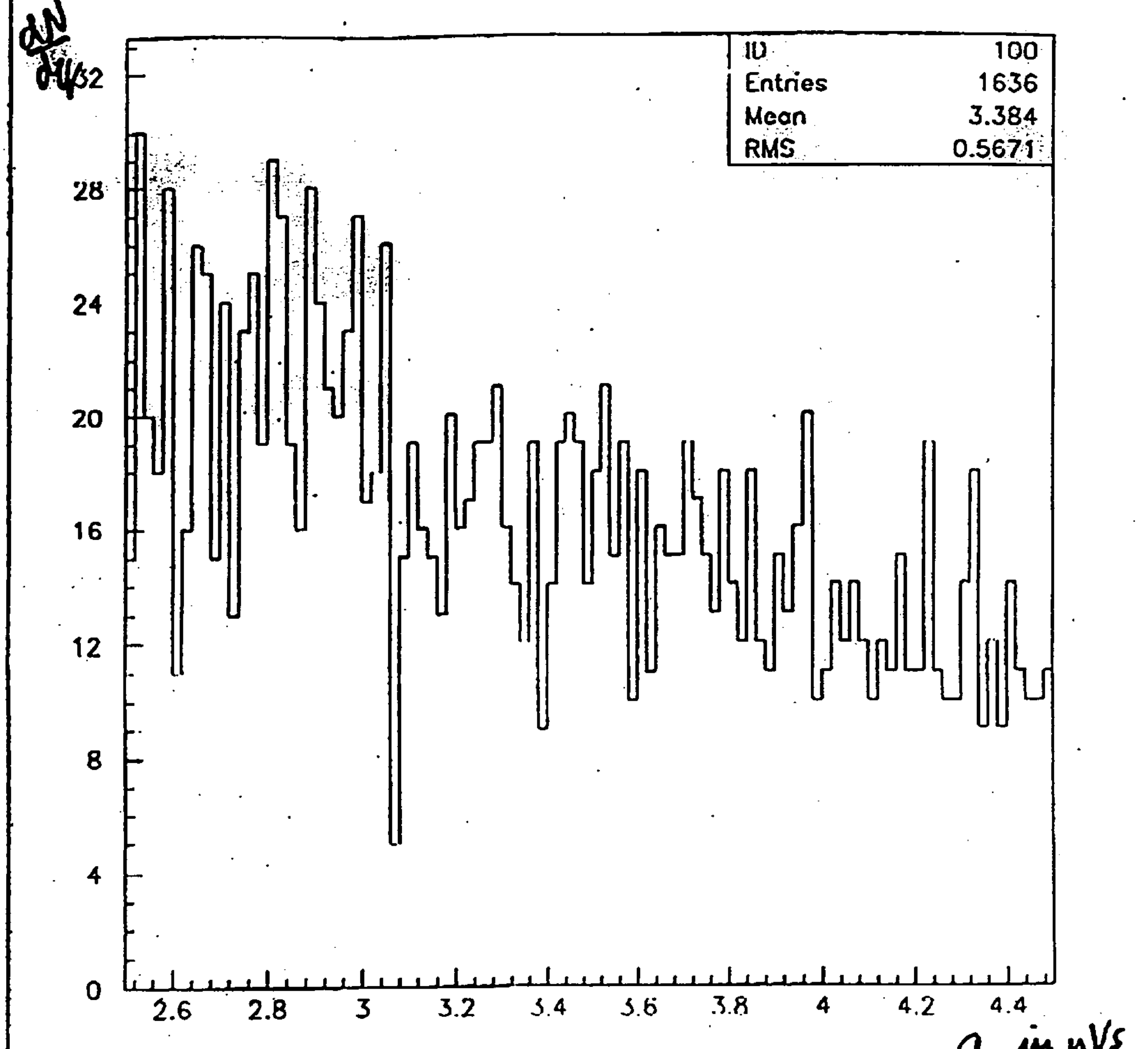
Rate = 1.7 Hz



Normal ^{228}Th Stop Pulse (α peak)



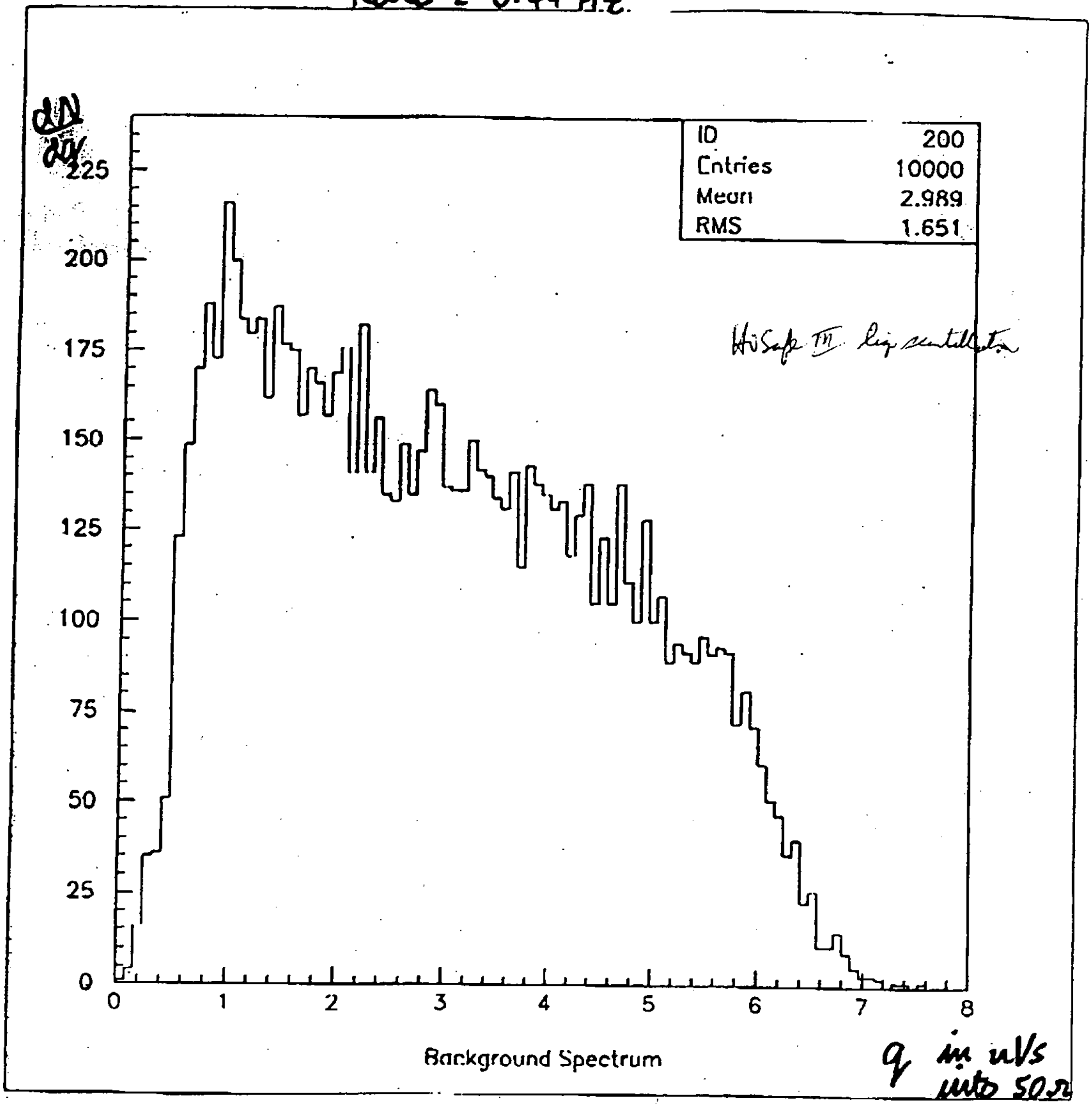
γ Background cut to region of α peak
Notice energy cut gives reduction of around 7



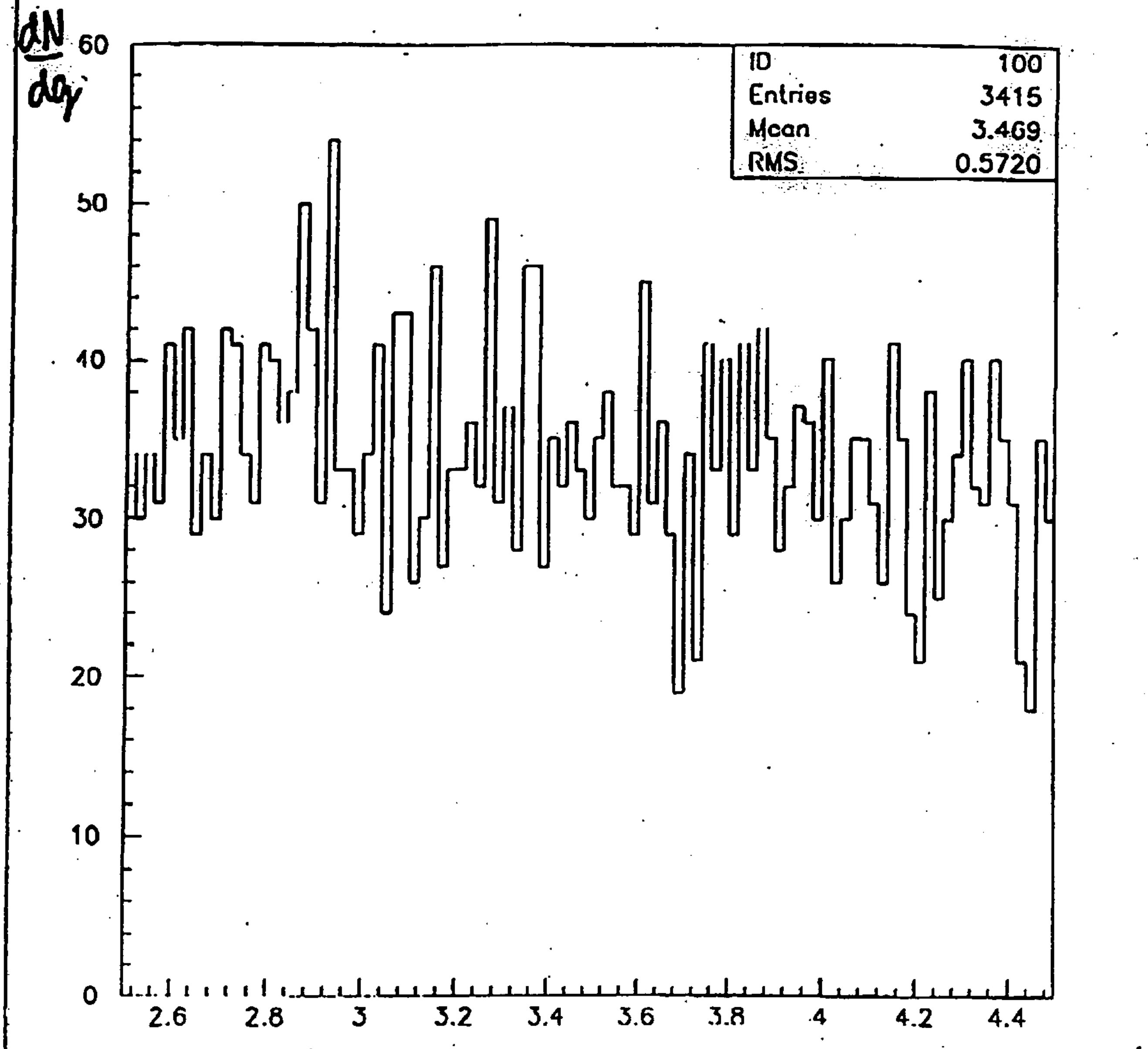
Background spectrum in alpha peak region γ Background spectrum in COPPER + STEEL shielding system

exterior
through Pb, Cu

Rate = 0.44 Hz.



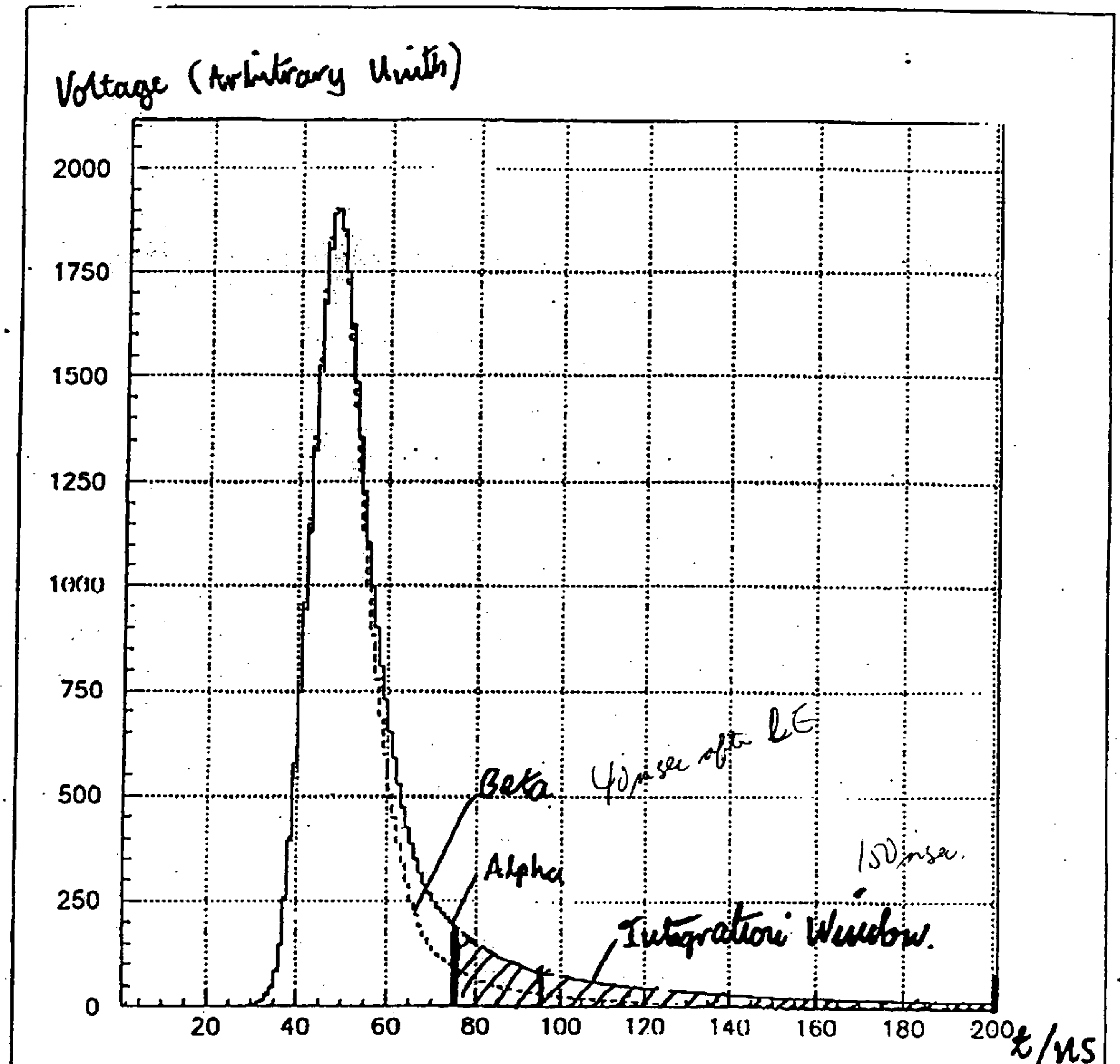
[illegible] copper + steel system. Hardening of spectrum leads to energy cut factor of only 3, but rate has reduced by a factor of 4, and this comes in squared in γ/γ rate.



Background spectrum in alpha peak region

Average Pulse Shapes for ~2000 α's & β's

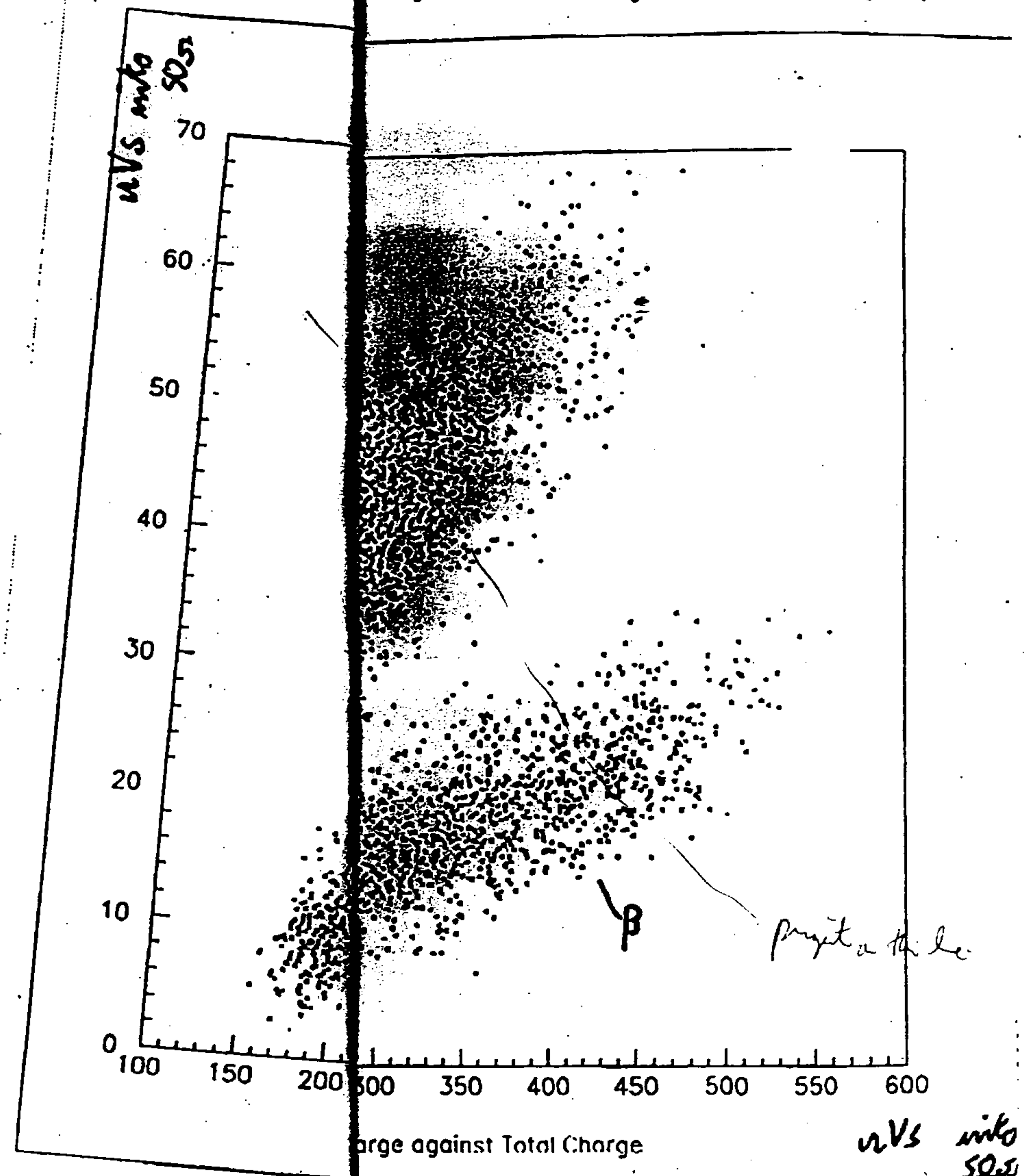
Pulses are normalised to peak height and shifted to cross at 10% peak height crossing.



Channel 35 is 10% crossing time

... separation of αs & βs

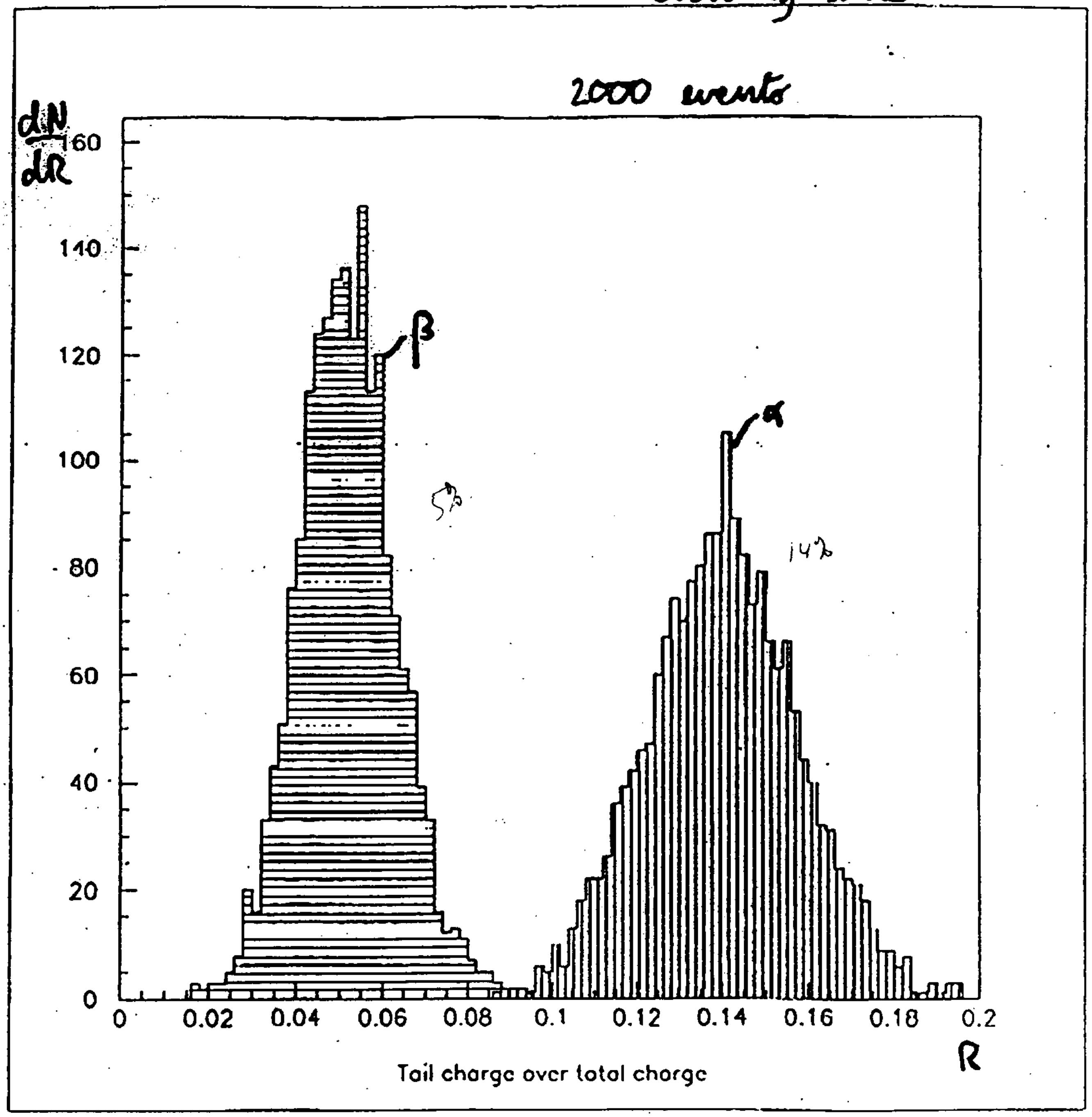
Tail calculated from 40us after 10% cross after 10% time. Total charge is from to 200us after 10% cross.



...arge against Total Charge

Tail charge is calculated from 40ns → 200ns after 10% crossing time

Total charge is from 0ns → 200ns after 10% crossing time



Ratio, R, of tail charge to peak voltage.
Tail charge is as given in the previous figure
Notice larger separation of mean's, but also
larger widths of the distributions.