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LUCAS HEIGHTS RESEARCH LABORATORIES

MEASUREMENT OF THE RESPONSES OF SMALL LITHIUM GLASS SCINTILLATORS TO PROTONS, DEUTERONS AND ALPHA PARTICLES

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ABSTRACT

The application of lithium glass detectors to the measurement of the tritium production rate distribution in fusion blanket assemblies is under investigation at Lucas Heights. As a consequence of neutron-induced reactions within the glass detector, many charged particles are produced with energies ranging from zero to about 20 MeV, all of which produce scintillations. To isolate the signals arising from the tritium production reactions $^{6}Li(n,\alpha)T$ and $^{7}Li(n,n'\alpha)T$ within the glass, the characteristic responses of the glass to all charged particles are required.

As part of this investigation, the response characteristics of thin lithium glass scintillators to protons, deuterons and alphas have been measured as a function of particle energy (0.8 to 2.7 MeV), lithium concentration (2.4 to 8.3 wt %) and lithium enrichment (95 wt % ^{6}Li to 99.99 wt % ^{7}Li).

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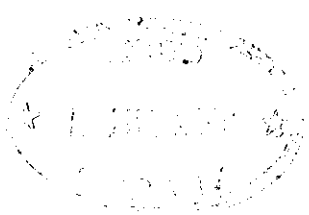

1. INTRODUCTION

Silicate glasses containing the oxides of low mass elements such as lithium, aluminium or magnesium, in combination with cerium oxide (the activator), emit light flashes when traversed by energetic charged particles, lithium glasses producing the maximum light output at a given particle energy [Anderson *et al.* 1960, 1961]. The scintillations produced in lithium glass by neutrons are due to the energetic charged particles emitted in the reactions

$$^{6}Li + n = T + {}^{4}He + 4.8\ MeV; \quad (1)$$

$$^{7}Li + n = T + {}^{4}He + n' - 2.8\ MeV. \quad (2)$$

Since these reactions are used for tritium breeding in the fuel cycle of deuterium-tritium fusion reactors, lithium glass detectors can also be used, in principle, to measure the rate of formation of tritium in an experimental fusion blanket assembly.

The energy-dependent cross sections of **reactions 1** and **2** are shown in **figure 1**. Below about 2 MeV, the cross-section for **reaction 1** is so large that even for low concentrations of ^{6}Li in the glass almost all neutron absorption corresponds to this reaction. Glasses, containing lithium enriched in ^{6}Li [Ginther 1960] are now well established for thermal neutron detection.

Above about 4.5 MeV, the $^{6}Li(n,\alpha)T$ cross section falls to such a low value that tritium is produced predominantly from the $^{7}Li(n,n'\alpha)T$ reaction. In this energy region, however, contributions from electrons arising in gamma-ray interactions and charged particles from the many neutron-induced reactions in the glass become significant. To extract the signals associated with tritium production in the lithium isotopes from the total light output requires a knowledge of the response of glass scintillators to electrons and all relevant charged particle types as a function of their energy up to about 20 MeV.

Although lithium glass scintillators have been in use for more than 25 years, the only data found in the literature pertaining to the efficiency with which they convert the kinetic energy of heavy charged ions into scintillation energy were

(a) the tritons (2.73 MeV) and alpha particles (2.07 MeV) released in the $^{6}Li(n,\alpha)T$ reaction by thermal neutrons [Anderson *et al.* 1961, Firk *et al* 1961, Bollinger *et al.* 1962, Wraight *et al.* 1965, Spowart 1969, 1976, Jensen and Czirr 1983],

(b) the 5.48 MeV alpha particles from ^{241}Am [Spowart 1976], and

(c) the 5.15 MeV alpha particles from ^{239}Pu [Firk *et al.* 1961].

In all of these references it was acknowledged that the scintillation efficiency of lithium glass is greater for electrons than for heavier charged particles, and that for the latter the efficiency varies inversely with the ionisation density.

The measurement and analysis of the relative scintillation efficiencies of small lithium glasses, of different lithium content and enrichment, to protons, deuterons and alpha particles over an energy range 0.8 to 2.7 MeV is described. The objective of the investigation was to further the understanding of these responses and to contribute to the pool of data required for the development of detectors for tritium production measurements in future fusion blanket studies.

2. RESPONSE OF LITHIUM GLASSES IN A FUSION BLANKET ASSEMBLY

2.1 Interactions Producing Scintillations

The basic ingredients used in lithium glasses are the oxides of silicon, lithium, magnesium, aluminium, and cerium. Except for cerium oxide, which has a concentration of about 4 wt % in all glasses, the fractional content of each material depends on the amount of lithium included in the composition [Spowart 1976].

Over the range of gamma energies inherent to a fusion blanket assembly (about 30 keV to 20 MeV), Compton scattering is the predominant interaction mechanism within the lithium glasses, with scintillations arising from the scattered electrons. Within the neutron energy range (14 MeV to thermal), many nuclear interactions are possible with each of the materials in the glass. However, except for the $^{6}Li(n,\alpha)T$ reaction, all reactions which produce charged particles have thresholds at about 2 to 3 MeV. For neutrons with energy

greater than this, a variety of charged particles with a wide range of energies are produced by

(a) inelastic (n,n'γ) collisions which produce heavy recoil nuclei with only a fraction of the incident neutron energy;

(b) (n,p), (n,d), (n,t), (n,α), (n,pα), in which all of the relatively light reaction products have a non-zero electronic charge; and

(c) (n,γp), (n,n'p), *etc.* which produce both charged particles and one or more particles with no electronic charge.

Thus, even for monoenergetic incident neutrons, the light output from a glass scintillator will be a superposition of the signals arising from a number of different charged particles. To extract information relating to tritium production from the lithium isotopes in the high neutron energy region therefore requires a knowledge of the responses of the lithium glasses to the individual charged particles as a function of their energy.

2.2 Factors Affecting Scintillation Response

The scintillations observed in cerium-activated glass are initiated by dissipation of the kinetic energy of charged particles within its volume. The scintillation process in the glass is similar to that in the NaI(Tl) crystal except that the small size (~2 nm) and random orientation of the crystal structures within the 'amorphous' glass material lead to light pulses with significantly smaller amplitudes.

A small fraction S_i (the scintillation efficiency) of the energy E_i of each charged particle 'i' is transmitted to the cerium activation centres (Ce_2O_3) *via* the crystal lattices in which they reside. There, the energy is converted into X_i photons with a mean wavelength, λ_p, of 385 nm (energy E_p of 3.14 eV) by the deactivation of the excited cerium atoms. The light energy emitted is

$$L_i = X_i\ E_p = S_i\ E_i \quad . \tag{3}$$

The energy transfer to the separate cerium activation centres is so rapid (10^{-6}s) that even when more than one charged particle is produced in a reaction, the light emitted is seen as a single pulse, *i.e.* the sum of the light signals from the individual charged particles.

2.3 Amplitude of the Observed Scintillation Pulses

The total energy of the reaction products, E_t, emitted in an interaction is related to the incident neutron energy, E_n, by the following relationship:

$$E_t = E_n + Q \quad , \tag{4}$$

where Q is the mass difference between the initial and final particles involved in the reaction. Q may be positive (exothermic) or negative (endothermic). If, for example, two particles are emitted in the interaction, then

$$E_t = E_1 + E_2 \quad , \tag{5}$$

and the amplitude of the scintillation pulse will be

$$L_t = S_1\ E_1 + S_2\ E_2 \quad . \tag{6}$$

Although the total energy released in a specific interaction is uniquely dependent on the incident neutron energy and the reaction Q-value, the distribution of energy between the individual reaction products is variable, being determined by the angle at which they are emitted. Thus, if one of the emitted particles has no electronic charge, or if the scintillation efficiencies differ for the two charged particles, then the light output will vary non-linearly with the total energy released in the interaction. The same principles apply for interactions in which more than two particles are emitted.

2.4 Detection of the Emitted Light Signals

Experimentally, the light output pulses from the glass scintillator are collected and amplified using a photomultiplier tube. The anode pulses of the latter are sorted on the basis of their amplitudes and displayed as a pulse height spectrum on the video screen of a multichannel analyser (MCA). The channel number is therefore proportional to the light energy emitted in individual interactions within the glass phosphor.

Some of the photons originating in the glass are lost in transmission between the phosphor and the photocathode so that only a fraction G eventually impinges on the latter. At the photocathode, the photons are

converted to electrons with an efficiency Q, and a fraction g of the latter (the electron-optical focusing efficiency) is collected by the first dynode. These electrons are then multiplied at n successive dynodes, producing an overall gain of M, so that the total number P collected at the anode is

$$P = A M \quad , \tag{7}$$

where

$$A = X T \quad , \tag{8}$$

$$T = G Q g \quad , \tag{9}$$

A is the number of electrons reaching the first dynode, and T is the probability that a scintillation photon created in the phosphor will produce an electron at the first dynode (the photon transfer efficiency).

The values of the various parameters in the above equations are dependent on the characteristics of the phosphor-photomultiplier combination. Statistical fluctuations in the various processes produce variations in the amplitude, P, of the anode pulses for ionising radiations of fixed incident energy. The distribution of these pulse amplitudes is principally determined by the variance of the electron distribution arriving at the first dynode. For 'A' greater than about 20, both this distribution and that of the anode pulses closely approximate to a Gaussian shape. From an analysis of the statistics of the processes involved, Breitenberger [1955] showed that the relative variance, v(P), of the measured pulse height distribution of the anode signals is given by

$$v(P) = \frac{1 + v(M)}{x T} \quad , \tag{10}$$

and that the relative variance v(M) introduced by the electron multiplication processes along the dynode chain is given by

$$v(M) = \frac{1}{(R - 1)} \quad , \tag{11}$$

where R is the mean stage gain between the n successive dynodes and is related to the overall gain by $M = R^n$.

For a pulse height distribution which is Gaussian in shape, the variance v(P) is related to the resolution η (defined as the full width of the distribution at half its maximum intensity divided by the mean) by the relation

$$\eta^2 = 5.545\, v(P) \quad . \tag{12}$$

Combining the above equations for the response of the glass scintillator to a single charged particle i with kinetic energy E_i , produces an expression for its scintillation efficiency:

$$S_i = \frac{1}{G Q g} \frac{R}{R-1} \frac{5.545}{\eta^2} \frac{E_p}{E_i} \quad . \tag{13}$$

3. EXPERIMENTAL DETAILS

3.1 Procedure

The glass scintillators listed in **table 1** were irradiated in turn by protons, deuterons and alpha particles each of 0.8, 1.2, 1.7, 2.2 and 2.7 MeV, a thin self-supporting gold foil being used to back-scatter the particles from the beam of the AAEC's 3 MV Van de Graaff accelerator into the scintillators. Pulse height spectra from the light output were recorded over intervals of 500 seconds, the same photomultiplier (PM) tube and data acquisition system (DAS) being used to provide an intercalibration between all measurements. Normalisation of all results to a common datum was obtained from energy calibrations of the MCA, during the irradiation of each glass with each charged particle type, using a range of standard gamma sources (^{22}Na, ^{24}Na, ^{54}Mn and ^{137}Cs).

3.2 Detection System

The glass scintillators listed in **table 1** constitute the full range of lithium compositions available from Nuclear Enterprises Ltd, Edinburgh, Scotland. Each scintillator contained about 4 wt% of Ce_2O_3. All were in the form of right cylindrical sections, 12.7 mm diameter and 3mm thick. For uniformity of response, glasses without surface reflector coatings were used [Spowart 1969].

Because of the very small range traversed by the charged particles in solid material, it was necessary that they enter the glass scintillators directly within the vacuum system of the beam tube. The glasses were mounted over collimator holes in one of the scattering ports of the target chamber, a vacuum seal being

provided by an O-ring concentric with the collimator. The end window of the PM tube was then sealed to the glass scintillator using high vacuum silicone grease. The PM and DAS are described in **appendix A**.

3.3 Target Chamber

The target chamber, shown in **figure 2**, was used to house the gold scattering foil and two glass scintillator-photomultipler detection systems. The gold foil was supported on a brass disc on the central axis of the chamber set at 90° to the beam direction. The area of foil presented to the beam was defined by a 10 mm diameter hole in the centre of the support disc. The energy spread produced by the gold foil was 40, 10 and 3 keV, respectively, for the alpha particles, deuterons and protons arriving at the scintillator.

The two scattering legs of the chamber were terminated by flange plates on which the glasses were mounted. Holes of diameter 4 mm were provided at the centre of each plate to collimate scattered charged particles entering the front surface of the glass scintillators. The distance between the gold foil and front surface of the glass was 210 mm. The accelerator beam, whose area of 2 mm diameter is defined by a collimator upstream of the chamber, entered the target chamber *via* the tube positioned between the two scattering legs.

3.4 Measurement of the Scintillation Efficiencies

The relative efficiencies with which the glass scintillators converted the energy of the charged particles into light energy were determined by locating the channel in which the maximum intensity occurred in the pulse height spectrum for each response measurement. These were converted to equivalent electron energies using the calibrations obtained with the standard gamma sources. Absolute scintillation efficiencies were obtained by normalising all results to the absolute value of 1.52 per cent for electron interaction in an identical 3 mm thick NE905 glass scintillator, derived from the measurements reported by Spowart [1969, 1970].

4. RESULTS AND DISCUSSION

4.1 Variation of Scintillation Response with Particle Energy

The calibration of the MCA scale in terms of electron energy, obtained using the standard gamma sources, is shown in **figure 3** for the NE902 glass scintillator. The light output of all glasses varied linearly with electron energy and their relative scintillation efficiencies were determined by linear least squares fitting. The absolute efficiencies of all glasses are given in **table 1**.

For the heavier charged particles, the light output increased non-linearly with energy, the relative variation depending on both glass composition and particle type. The pulse height distribution of the light output of the NE902 glass for 2.1 MeV protons is shown in **figure 4**. Relative responses of the NE902 glass scintillator to the different charged particles as a function of their kinetic energy are illustrated in **figure 5**. Normalisation of all measured data to those obtained for protons with the NE902 glass revealed that variation with energy for all glasses and all three particle types conformed to the same general shape. For the equivalent electron energy L_i (MeV) of the light output for a charged particle of kinetic energy E_i (MeV), least squares fits to the data produced a common equation of the form

$$L_i = 0.74\, E_i\, (1 + 0.35\, E_i)/k \quad , \tag{14}$$

where k is the normalisation factor for particle and glass type. Values of k are listed in **table 2** for all glasses.

From **table 2** it can be seen that the response varied inversely with the ionisation density of the particle. This effect, which is observed in all types of scintillator [Birks 1964], is attributed to 'ionisation quenching', which results from the higher specific energy losses for heavier particles and higher electronic charges. It is possible that both the non-linearity of responses and lower efficiency for more heavily ionising particles could arise as a consequence of the very short range of the ionising particles coupled with a lower scintillation efficiency near the edge of the glass and/or loss of photons from the unreflected surface.

For the excitation of scintillations, the ionising particle must deposit energy in the crystal lattice in which the cerium activator resides. The average size of such lattices is about 2 nm [Spowart 1976] and their axes are randomly oriented. There is no evidence to indicate that either the size or the orientation of these lattices is any different at the surface to that in the interior. Hence, because the range of even the lowest energy alpha particles exceeded 2 nm, the statistical effects produced by the particle irradiation at the surface of the glass should be similar to those produced by particles distributed throughout its volume.

The light from each activation centre is emitted isotropically [Spowart 1970], hence the number of photons emitted in the forward direction is independent of the depth below the glass surface at which they are produced, as is the loss of light from the unreflected surface of the glass scintillator. There will therefore be no dependence of the light collected by the PM on the particle energy resulting from this effect.

That similar differences have been observed between the responses of charged particles absorbed throughout the volume of the glass, *e.g.* the alpha particle and triton emitted in thermal neutron capture by ^{6}Li (see discussion in **section 4.3**), rules out an explanation for the observed variations with ionising particle density based on the above postulated edge effects.

4.2 Variation of Scintillation Response with Lithium Content

The relative variations of scintillation responses of the glasses with lithium content are shown in **figure 6** for electrons and **figure 7** for the heavier charged particles. The present results indicate that, for each type of charged particle, the amplitude of the light output decreases with increase in lithium content. Within the statistical errors of the measurements, the variations of the normalised scintillation responses with lithium content were the same for all particles, including electrons. Although significant differences were observed between glasses containing different lithium enrichments (but with the same total lithium content), no systematic variation in light output with lithium enrichment could be established.

To convert the relative scintillation efficiencies to the absolute values given in **table 1** and **figures 6 and 7**, results were normalised to the absolute value for electrons in a 3 mm NE905 glass reported by Spowart [1970]. The latter was consistent with an independent calibration at Lucas Heights based on **equation 3** and the measured values of the peaks and resolutions of the pulse height spectra produced by protons in the NE905 glass.

4.3 Comparison with Previous Investigations

Many investigations [Anderson *et al.* 1961; Kirk *et al* 1961; Bollinger *et al.* 1962; Wraight *et al.* 1965; Spowart 1969, 1970, 1976; Jensen and Czirr 1983] have shown that the scintillation produced in the glass by the combined kinetic energy of the triton (2.73 MeV) and alpha particle (2.05 MeV), emitted in the ^{6}Li (n,α)T reaction, is equivalent to that of an electron in the range 1.5 to 1.6 MeV. A value of 1.5 was obtained in the present investigation. From extrapolations of their ^{239}Pu and ^{241}Am alpha particle measurements [Bollinger *et al.* 1961; Firk *et al.* 1961 and Spowart 1976], ratios of the scintillation responses of the 2.73 MeV triton to the 2.05 MeV alpha particle were estimated to be 4, 3 and 5 respectively; a value of 4.5 was found for the present results.

The agreement obtained between the results of the present investigation and data reported in the literature provides good confirmation of the derived empirical variation of the glass responses with particle type and energy. The results thus constitute a valuable contribution to the data required for assessing the feasibility of using lithium glass detectors for tritium production measurements in fusion breeder blanket experiments.

5. CONCLUSIONS

The light signals produced in glass scintillators by energetic charged particles have been shown to vary with the lithium content of the glass and the energy, mass and electronic charge of the detected particle. The relationship between the amplitude of the light signals and the kinetic energy transferred to the glass by electrons is linear, whereas for the much heavier protons, deuterons and alphas it is non-linear, the shape of the non-linear curves being the same for all the heavy particles and for all glasses studied.

For all charged particles studied, the light output of the scintillators decreased with increase in the lithium content of the glass; within the statistical errors of the measurements, the shape of this relationship appeared to be the same for all particles, including electrons. For all glasses, the spread in the normalisation factors for the different charged particles increased with increase in lithium content.

The scintillation efficiency of all the glasses was greatest for electrons and least for alpha particles. For kinetic energy transfers of 1 MeV, the mean response of all glasses to electrons exceeded that due to protons, deuterons and alpha particles by factors 2.1, 2.8, and 10.3, respectively.

6. ACKNOWLEDGEMENTS

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TABLE 1
DETAILS OF LITHIUM GLASS SCINTILLATORS

Glass Type	Lithium Content Total Lithium (wt%)	Lithium Content $>^{7}$Li/Li (wt%)	Scintillation Efficiency (%)
NE901	2.40	natural	2.17
NE902	2.26	5.00	1.90
NE903	2.41	99.99	1.91
NE904	6.60	natural	1.95
NE905	6.23	5.00	1.52
NE906	6.63	9999	1.44
NE907	7.50	natural	1.18
NE908	7.08	5.00	1.01
NE909	7.54	99.99	1.08
NE912	7.80	5.00	1.02
NE913	8.30	99.99	0.82

TABLE 2
NORMALISATION FACTORS FOR EQUIVALENT ELECTRON ENERGY

Glass Type	Normalisation Factors Protons	Deuterons	Alphas
NE901	2.93	2.85	9.33
NE902	2.10	2.80	9.50
NE903	2.00	3.00	9.53
NE904	2.18	2.80	9.24
NE905	2.05	2.72	10.23
NE906	2.14	2.69	9.88
NE907	2.09	2.96	11.53
NE908	1.91	2.84	10.22
NE909	2.10	2.59	11.88
NE912	2.11	2.91	11.43
NE913	1.89	2.65	10.54
Mean	2.05	2.80	10.28

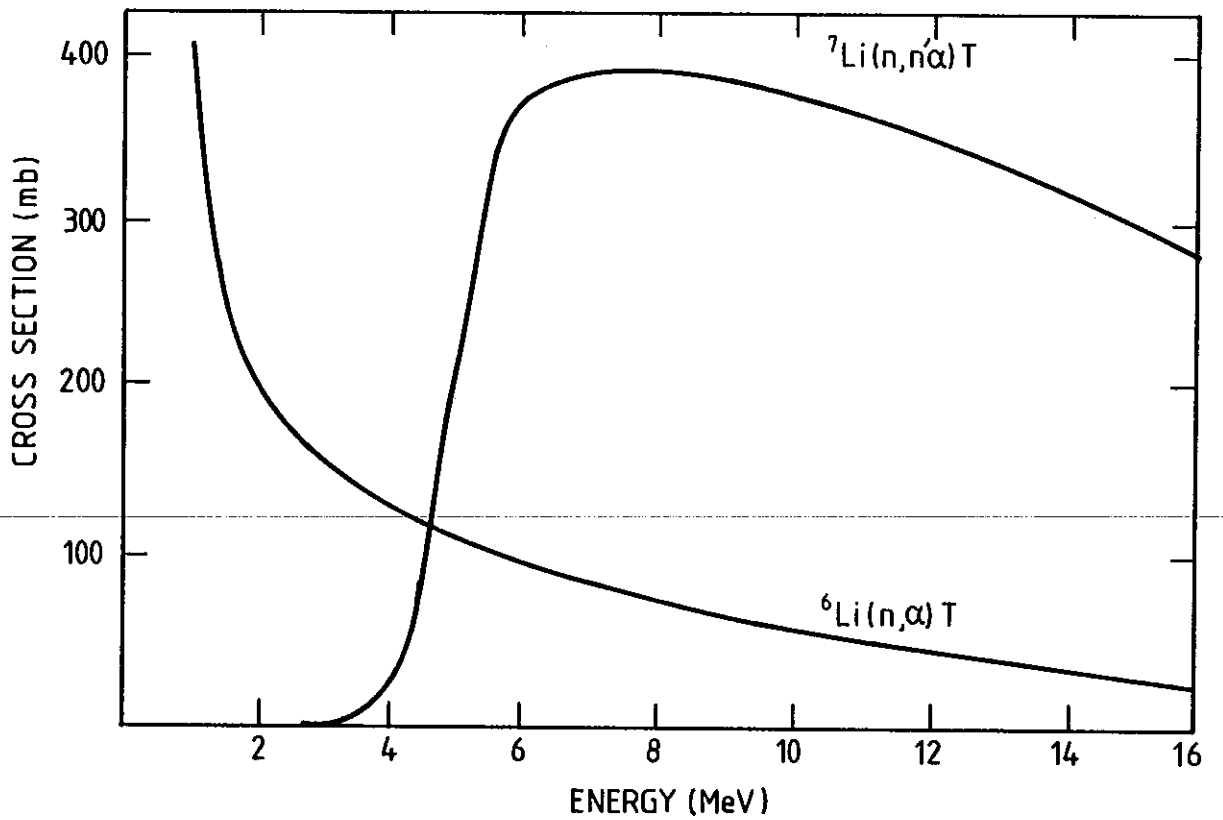

Figure 1 Variation of Tritium Production Cross Sections

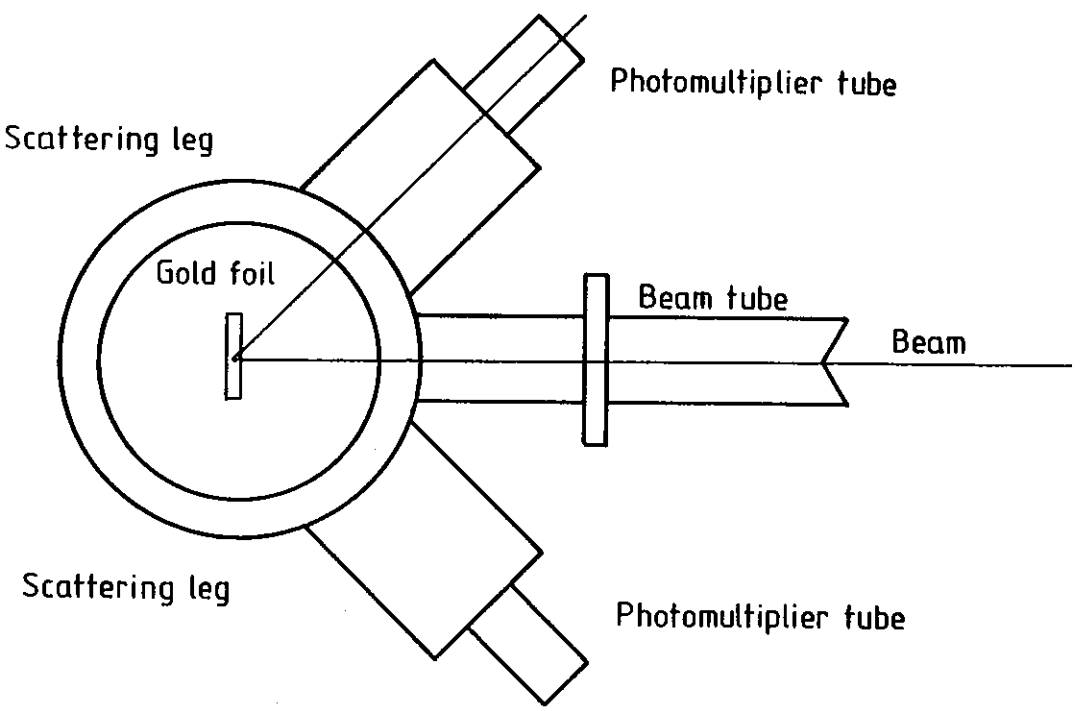

Figure 2 Plan of Target Chamber

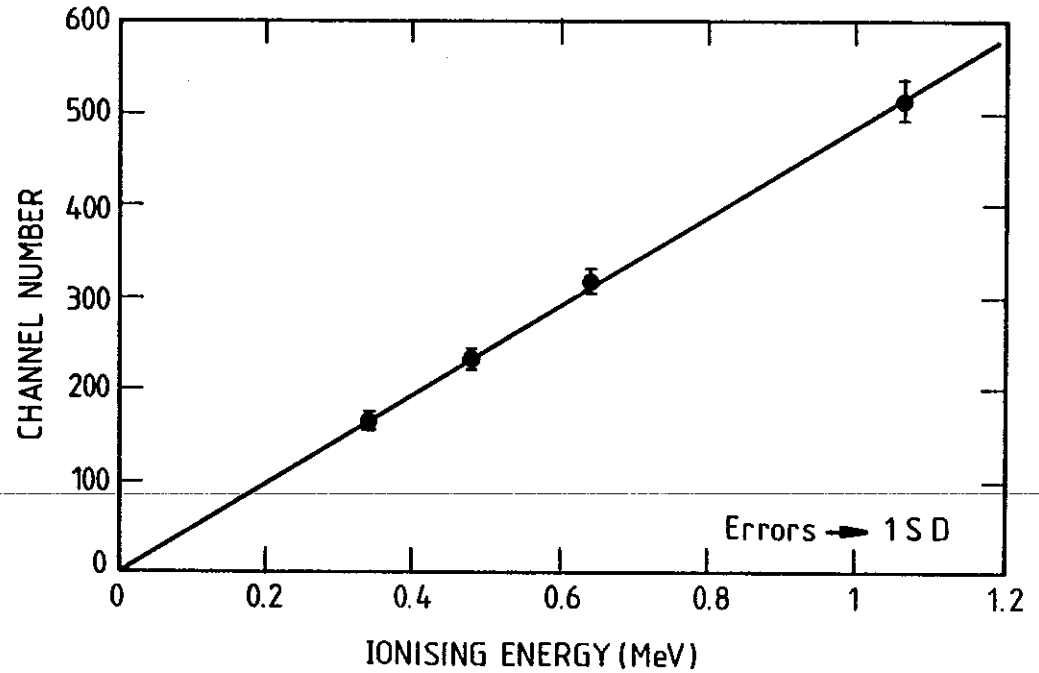

Figure 3 Variation of Light Output with Electron Energy

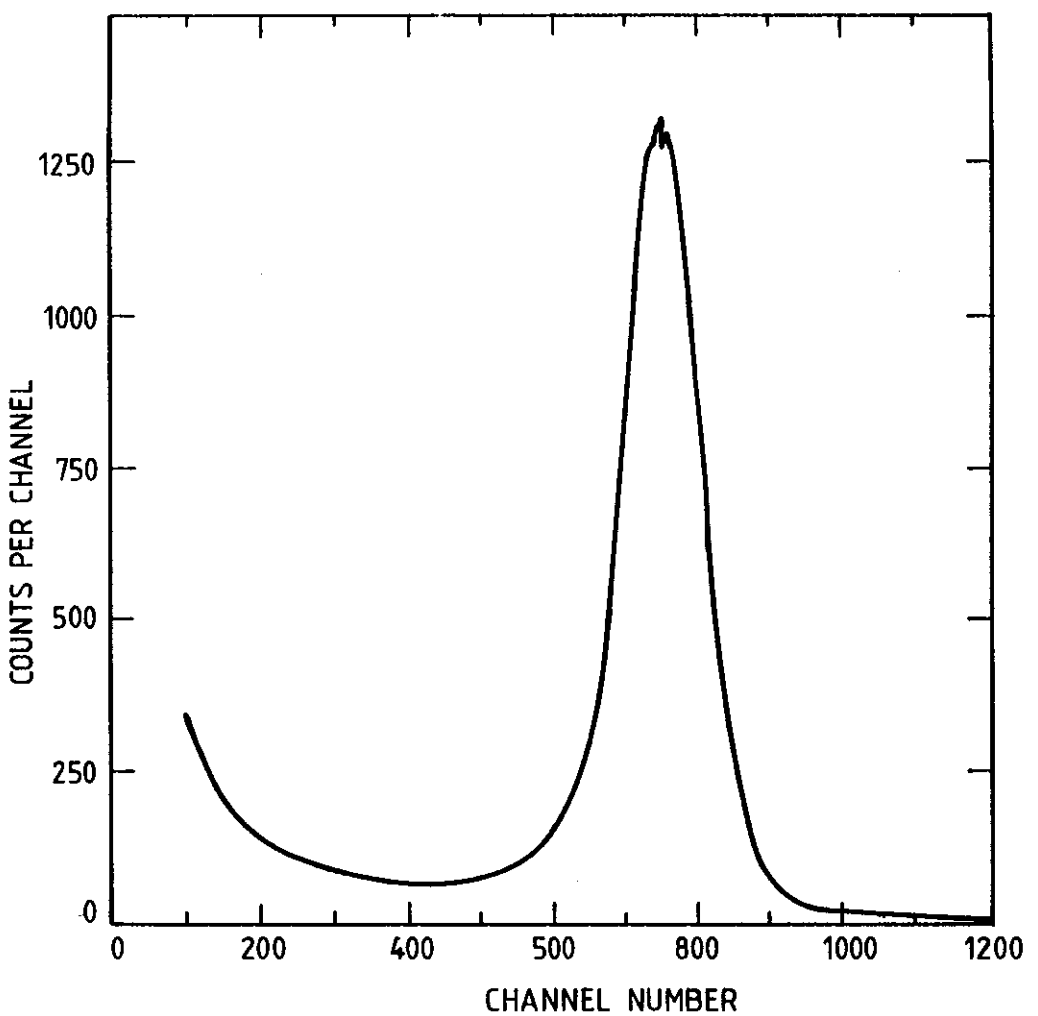

Figure 4 Pulse Height Spectrum for Protons in the NE902 Glass

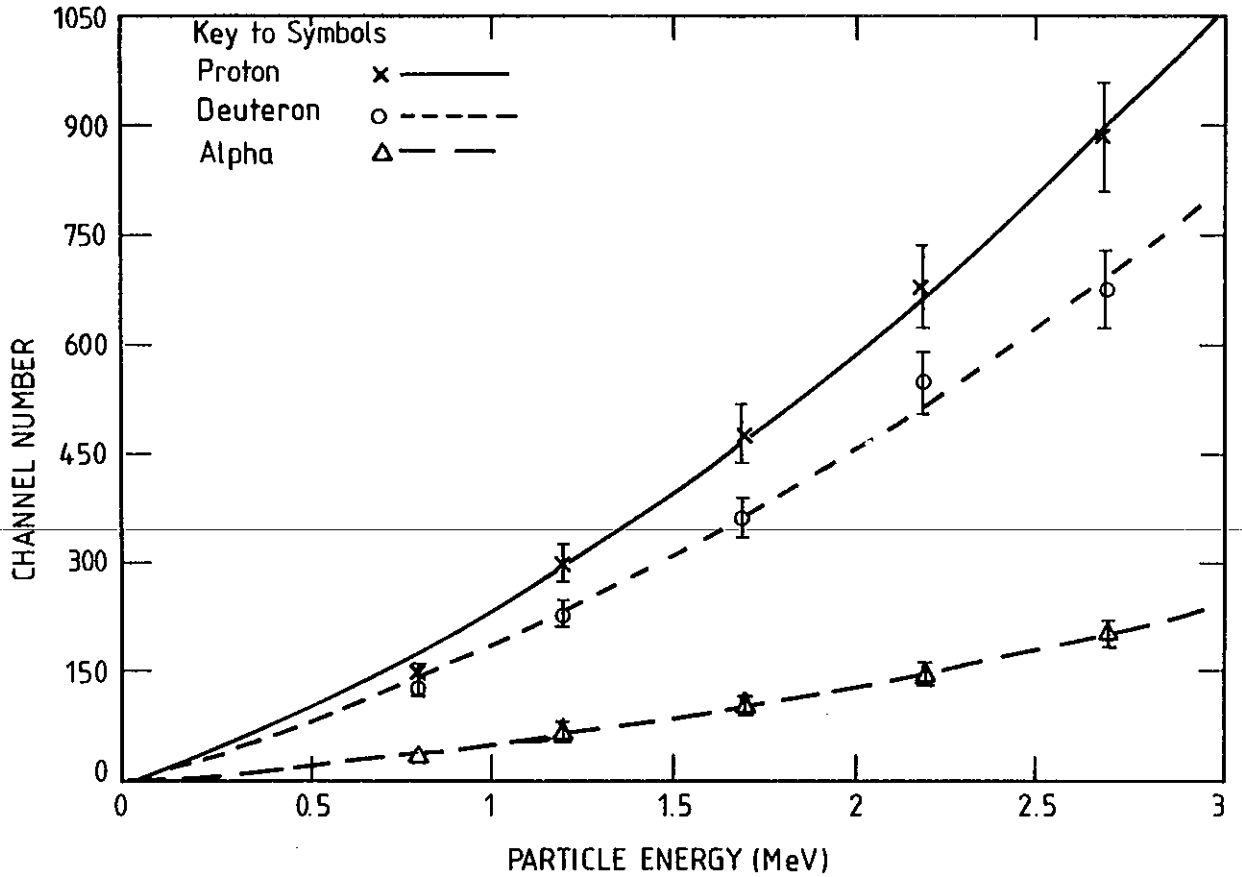

Figure 5 Charged Particle Responses for the NE902 Glass

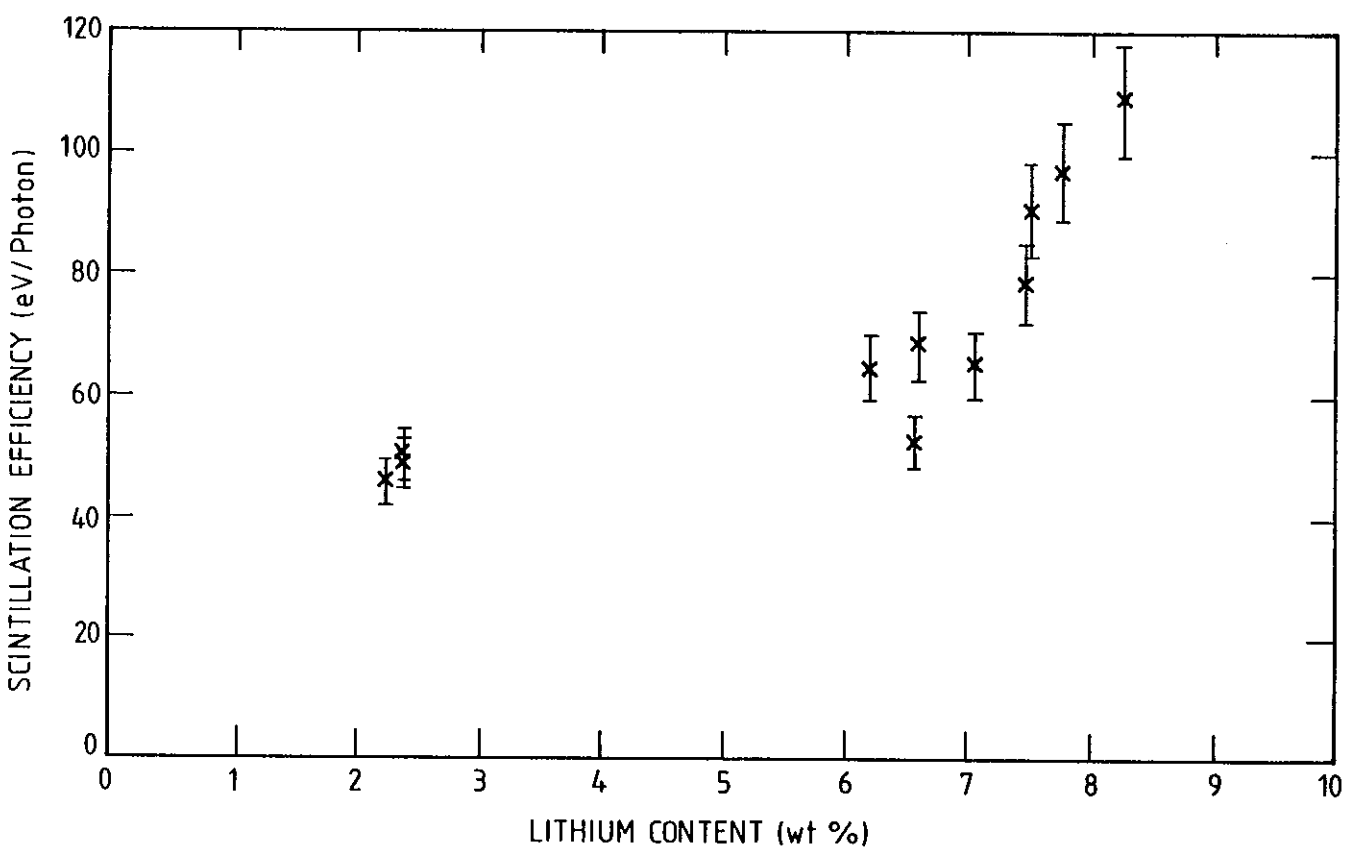

Figure 6 Variation of Light Output with Lithium Content (Electrons)

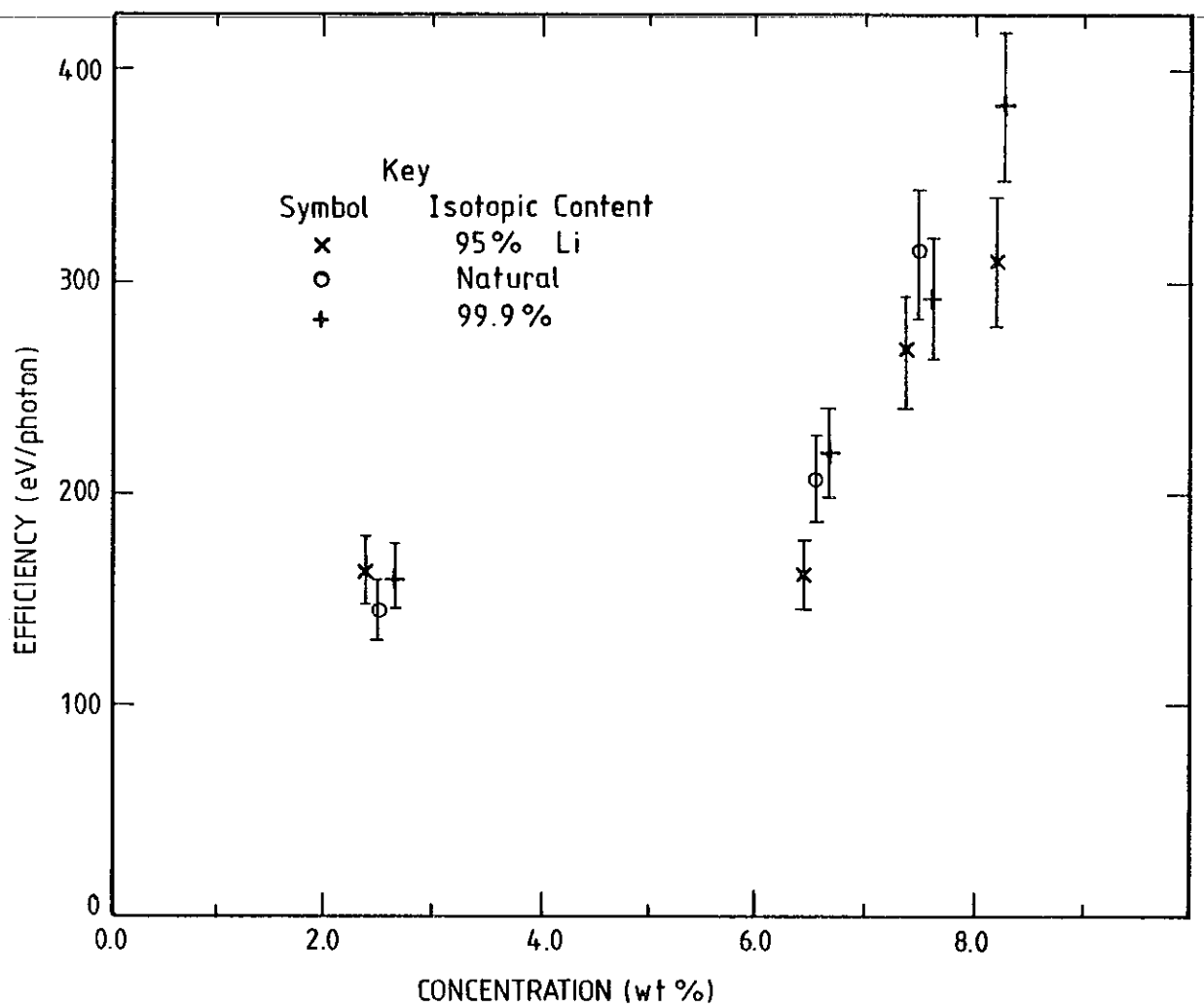

Figure 7 Variation of Light Output with Lithium Content (Protons)

APPENDIX A
DESCRIPTION OF PHOTOMULTIPLIER AND DATA ACQUISITION SYSTEM

A1 PHOTOMULTIPLIER

The data reported here were acquired with a Philips PM tube type PM1911. The tube has a flat end window, a 14 mm semitransparent S11 photocathode, 10-stage linear focusing and a CuBe secondary emitting surface. It was coupled to a specially constructed, high stability dynode chain connected to an Ortec 556 high voltage supply. To ensure optimum collection efficiency, the voltage between the cathode and first dynode was twice that between the other stages.

To ensure stability during operation, the overall voltage supplied to the dynode chain was set well below the maximum rating (-1750 V) at -1100 V. At the selected voltage, the tube had a quantum efficiency C of 15 per cent, an electro-optical efficiency g of 60 per cent, anode spectral sensitivity of 17 mA/W, a gain of 3×10^5 and a 'dark' current of less than 0.6 nA. To screen against the influence of stray magnetic fields, the tube was protected by a mumetal shield.

A2 DATA ACQUISITION SYSTEM

The signal from the anode of the PM was fed *via* a charge-sensitive pre-amplifier of time constant 50 μs, a spectroscopic amplifier (Ortec NIM model 2020) with a shaping time constant of 500 ns, and an analogue-to-digital converter (ADC, Canberra Instruments Series 40). For all measurements, a 1023 channel display was used, which allowed up to four pulse height spectra to be stored and inspected at any time during the measurements. For detailed analysis, the data were transferred as individual files to floppy disc storage using a SIRIUS microcomputer; software written by the author was used to process the archival data.

The voltage signal from a precision pulse generator (Berkeley Nucleonics Corporation model PB-4) was used to calibrate the scale of the MCA. This instrument can supply voltage pulses with a controlled variable amplitude and repetition rate (linearity and reproducibility of 50 ppm) and a range of rise and fall times. Voltage pulses in the range of 0 to 10 V, with a rise time of 50 ns and of decay of 50 μs were fed directly into the MCA *via* the ADC at a repetition rate of 250 Hz at intervals interspersed between the glass response measurements. The differential linearity was less than 0.07 per cent in all cases.