AAEC/E650

AAEC/E650

AUSTRALIAN ATOMIC ENERGY COMMISSION
RESEARCH ESTABLISHMENT

LUCAS HEIGHTS RESEARCH LABORATORIES

ABSOLUTE MEASUREMENT OF THE RESPONSES OF SMALL LITHIUM GLASS SCINTILLATORS TO GAMMA RADIATION

by

A.W. DALTON

APRIL 1987

ISBN 0 642 59857 6

AUSTRALIAN ATOMIC ENERGY COMMISSION
RESEARCH ESTABLISHMENT

LUCAS HEIGHTS RESEARCH LABORATORIES

ABSOLUTE MEASUREMENT OF THE RESPONSES OF SMALL LITHIUM GLASS SCINTILLATORS TO GAMMA RADIATION

---

by

A. W. Dalton

ABSTRACT

The absolute scintillation efficiency and intrinsic resolution of lithium glass scintillators for electron excitation have been determined over a range of electron energies, lithium concentrations and lithium enrichments. Measurements of these response characteristics form part of a study on the possible use of such glasses for the determination of tritium breeding in fusion reactor blanket experiments.

The measurements were undertaken to establish a basis for extracting the information relating to tritium production reactions from the background signals induced within the glass scintillators by the neutron/gamma fields of a fusion reactor blanket. Criteria for the selection of glasses most suitable for tritium breeding measurements are discussed in terms of their observed responses.

National Library of Australia card number and ISBN 0 642 59857 6

The following descriptors have been selected from the INIS Thesaurus to describe the subject content of this report for information retrieval purposes. For further details please refer to IAEA-INIS-12 (INIS: Manual for Indexing) and IAEA-INIS-13 (INIS: Thesaurus) published in Vienna by the International Atomic Energy Agency.

BREEDING BLANKETS; EFFICIENCY; ELECTRONS; EXPERIMENTAL DATA; GAMMA RADIATION; GLASS SCINTILLATORS; LITHIUM 6; LITHIUM 7; NEUTRON REACTIONS; RESOLUTION; THICKNESS; TRITIUM

EDITORIAL NOTE

From 27 April 1987, the Australian Atomic Energy Commission (AAEC) is replaced by Australian Nuclear Science and Technology Organisation (ANSTO). Serial numbers for reports with an issue date after April 1987 have the prefix ANSTO with no change of the symbol (E, M, S or C) or numbering sequence.

CONTENTS

1. INTRODUCTION

Lithium glass scintillators have been studied at Lucas Heights to determine their suitability for monitoring the tritium produced in experimental fusion reactor blanket assemblies [Dalton 1987].

It should, in principle, be possible to measure the production of tritium at low and intermediate neutron energies from the scintillations produced in a glass enriched in ^{6}Li *via* the reaction

$$^6\text{Li} + \text{n} = \text{T} + {}^4\text{He} + 4.8\ \text{MeV} \quad , \tag{1}$$

and at high neutron energies using those produced in a glass enriched in ^{7}Li *via* the reaction

$$^7\text{Li} + \text{n} = \text{T} + {}^4\text{He} + \text{n}' - 2.82\ \text{MeV} \quad . \tag{2}$$

However, as discussed in the earlier report, scintillations produced by charged particles and Compton electrons within the glass, arising respectively from competing reactions induced by the neutron and gamma fields within a fusion blanket, contribute such a high background that detailed theoretical and experimental analyses are required to extract the information relating to tritium production from the measured data.

For an analytical approach, the tritium production information would be obtained from the measured light energy spectra using unfolding techniques based on the response characteristics of the various charged particles produced within the glass. The data required include the responses to electrons, protons, deuterons, tritons, alpha particles and heavier ions over the energy range zero to about 18 MeV. The measurement of some of these data was reported by Dalton [1987]; however, significantly more data are required before such an analysis can be attempted.

Experimentally, information relating to the tritium production from **reaction 1** could be obtained using a background compensation method based on the differences in the responses of ^{6}Li and ^{7}Li glasses measured in the same locations within the fusion blanket assembly [Yamaguchi and Nakamura 1986]. For this technique, it is necessary to match the responses of two glass scintillators to eliminate all background interactions common to both. The theory and measurements required to achieve this are described below.

2. THEORY

2.1 Interactions of Charged Particle Reaction Products

Reactions induced in a glass scintillator by the neutron flux of a fusion blanket assembly produce many types of charged particles, each with a wide range of energies [Dalton 1987]. The ranges of charged particle types and energies produced depend on the chemical composition of the glass, that is, on the number and concentration of its constituent materials. For commercially available glasses [Dalton 1987], these vary markedly as illustrated in **table 1**. Hence if two glasses of different enrichment are to have matching responses to all competing reactions when located at the same position in the assembly (that is, exposed to the same neutron and gamma flux distributions), they must have the same chemical composition.

2.2 Interaction of Gamma-rays

Gamma rays with energies up to about 10 MeV are generated within a fusion blanket assembly during operation (**figure 1**). Over this energy range, Compton scattering is the predominant mechanism for gamma interaction within lithium glasses [Evans 1958]; the cross section, σ, is a maximum at zero energy and decreases monotonically with increase in the incident gamma energy [Davisson 1965].

For a gamma-ray energy of E_g, electrons with a continuous distribution of energy ranging from zero up to a maximum value E_c, known as the Compton edge, are produced within the scintillator:

$$E_c = \frac{2\,(E_g)^2}{(m_oc^2 + 2\,E_g)} \quad . \tag{3}$$

where m_o is mass of the electron and c the velocity of light. In this investigation, Compton edge electrons were chosen as the ionising radiation with which to determine the response characteristics required for matching the performance of glass scintillators.

The probability that a Compton interaction will occur and the energy of the scattered electron be dissipated within the glass depend on the physical dimensions of the latter. For a glass scintillator irradiated by the isotropic flux found within a fusion blanket assembly, the important parameter for both effects is its mean chord length, given by R = 4V/S [Case *et al.* 1953]. In terms of this parameter, the fraction of incident gamma photons, f, involved in Compton interactions within the glass is

$$f = 1 - \exp(-\lambda_C R) \quad , \tag{4}$$

where $\lambda_C = NZ\sigma$ (the macroscopic cross section) and N and Z are, respectively, the mean atomic density and number of the glass material. Over the range of glasses used in the investigation, the value of NZ varied insignificantly, the mean being $(7.40 \pm 0.05) \times 10^{23}$.

The energy E_R(MeV) at which the extrapolated range of the electrons becomes equal to the mean chord length R (mm) is [Katz and Penfold 1952]

$$E_R = (0.355\, R)^{0.58} \quad . \tag{5}$$

For higher electron energies, the average energy deposited in the glass will be limited to E_R.

Glass scintillators of small dimensions were therefore selected because of their lower sensitivity to gamma radiation and to limit the maximum electron energy deposition. Their diameter (12.7 mm) was determined by the smallest commercially available photomultiplier (PM) suitable for this type of work; parameters for three glass thicknesses are given in **table 2**, which shows that both the gamma detection efficiency and the maximum electron energy transfer increase with glass volume for gammas of all energies.

2.3 Conversion of Ionising Energy to Light Energy

The absolute scintillation efficiency, S, with which the kinetic energy, E_i, of the ionising radiation is converted into light energy is

$$S = X\, h\nu / E_i \quad , \tag{6}$$

where X is the absolute number of photons of frequency ν per ionising event produced within the glass.

When a scintillator is coupled to a PM tube, the absolute numbers of electrons, A, collected on the anode per ionising event in the glass is

$$A = X\,T\,Q\,M \quad , \qquad (7$$

where T is the fraction of the original photons which reaches the photocathode, Q is the quantum efficiency o the photocathode for conversion of photons to electrons, and M is the overall gain produced by the PM tube.

Statistical fluctuations produce a spread in the number of electrons arriving at the anode which can b expressed in terms of the resolution η(A) of their distribution (full width at half maximum height divided by th mean). It has been shown that η(A) can be expressed in terms of two independent components [Garlick an Wright 1952; Breitenberger 1955];

$$\eta^2(A) = \eta^2(\mathrm{I}) + \eta^2(L) \quad , \qquad (8$$

where η(I) arises from variations (caused by local inhomogeneities in the glass) in the number of electron reaching the anode for a fixed quantity of light, X, released within the glass, and is characteristic of th composition and geometry of the glass; and η(L) arises as a consequence of the combined statistic fluctuations from all of the processes represented in **equation 7**. From the statistical analysis of Presco [1963], it can be shown that

$$\eta^2(L) = \frac{11}{X\,Q\,T} \quad . \qquad (9$$

On the basis of **equations 7** to **9**, a plot of the measured values of $\eta^2(A)$ against 1/A for a particular glas produces a straight line whose intercept on the ordinate axis at 1/A = 0 corresponds to the value of $\eta^2($ characteristic of the glass [Bisi and Zappa 1958]. Thus for each glass, values of η(L) can be derived fro **equation 8** for all other values of A. The light energy transferred from the glass scintillator to the photocathod can subsequently be derived from **equation 9** in absolute terms with Q equal to 15 per cent, the valu corresponding to the mean frequency (285 nm) of the light photons emitted by the glass scintillators, and T ≈ for the reflected surfaces used.

3. EXPERIMENTAL DETAILS

3.1 Equipment

The glass scintillators used in the investigation (**table 1**) were in the form of right cylindrical sections of diameter 12.7 mm and thickness 3 mm, with alpha alumina surface reflector coatings [Spowart 1969]. The glasses were mounted in turn directly onto the end window of a Philips type 1911 PM tube, using high vacuum silicone grease. Electrons with maximum energies ranging from 0.3 to 2.5 MeV were produced in the glasses by the Compton interactions of gammas emitted by calibrated ^{22}Na, ^{24}Na, ^{54}Mn and ^{137}Cs sources. The ^{24}Na source was prepared at Lucas Heights and the rest were obtained from the International Atomic Energy Agency laboratory at Seibersdorf, Austria.

The signals from the anode of the PM were fed *via* an AAEC pre-amplifier and an Ortec NIM model 2020 spectroscopic amplifier to a Canberra Instruments series 40 multichannel analyser (MCA). The scintillations produced by each gamma source were recorded by the MCA for each glass over a period of 1000 seconds. A typical energy spectrum is illustrated in **figure 2**. All data were transferred to a microcomputer where they were analysed using software written by the author. Voltage signals from a precision pulse generator (Berkeley Nucleonics Corporation model PB-4) were used throughout the investigation to maintain the same calibration for all measurements.

3.2 Determination of Scintillation Efficiency

The channel numbers (Z) of the MCA display are proportional to the electron pulses produced at the anode (A) hence $\eta(Z)$ is, by definition, equal to $\eta(A)$. The peak channel, Z_c, and resolution, $\eta(Z_c)$, were determined from each measured pulse height spectrum using the empirical criterion of Flynn *et al.* [1964]:

$$Z_c = 0.96\, Z_{50} \quad \text{, and} \tag{10}$$

$$\eta(Z_c) = (Z_{12.5} - Z_{87.5})/Z_c \quad , \tag{11}$$

where the subscripted Z values represents the channel numbers in which the intensities corresponded to 12.5, 87.5 and 50 per cent of the maximum, respectively.

The relative scintillation efficiencies, S, of the glasses were determined from the slopes of the lines obtained by plotting Z_c against E_c (*e.g.* **figure 3**). Their intrinsic resolutions $\eta(I)$ were derived from the intercepts, on the ordinate axis at $1/Z_c = 0$, of the lines obtained by plotting $\eta^2(Z_c)$ against $1/Z_c$. Calibration was maintained throughout the measurements to ensure that the scintillation efficiencies of all glasses were normalised to a common datum.

The values of $\eta(L)$ derived from these data (**equation 8**) and their relationship to the absolute number of light photons X incident on the photocathode (**equation 9**) provided an absolute calibration of the channel scale of the MCA in terms of light energy, hence the value of the constant k in

$$X = k\, Z_c \,. \tag{12}$$

All responses were converted to absolute values using this relation. The calibration was confirmed by a direct measurement of the pulse height distribution of the fixed amplitude light pulses emitted by a GaAs diode ($T = 1$ and $\eta(I) = 0$) [Whittlestone 1979].

4. RESULTS AND DISCUSSION

The light output from all glasses increased linearly with electron energy. This indicated that the scintillation efficiencies were independent of the latter and could be obtained from linear least-squares analyses of the measured data. The responses for the most efficient glass, NE902, are shown in **figure 3**; the absolute scintillation efficiencies and intrinsic resolutions of all glasses are listed in **table 3** (the errors in these estimates were about 8 per cent).

The absolute scintillation efficiency, S, is plotted as a function of lithium content in **figure 4**. It can be seen that the variations in S produced by changes in total lithium content differ in both form and quantity from those produced by changes in lithium enrichment (for constant lithium content). There is a correlation between the values of S and lithium content, with S decreasing by a factor of about 2.5 for a three-fold increase in lithium content. Although variations up to a maximum of about 30 per cent were obtained for a seven-fold increase in ^{7}Li enrichment (for fixed total lithium content), a systematic relationship was not observed. The difference between the pair of glasses of maximum and minimum ^{7}Li enrichment was, however, least for the lowest lithium content (NE902 and NE903) and greatest for the highest content (NE912 and NE913). Trends similar to those for S were observed for the variation of $\eta(I)$ with lithium content and enrichment although the relative magnitudes of all changes in the latter were smaller.

The absolute scintillation efficiency of an NE905 glass has been determined for the ionisation energy (4.8 MeV) produced in the ^{6}Li (n,α)T reaction by thermal neutrons [Spowart 1969]. Combining this result with the relative responses for electron excitation of the same NE905 glass [Spowart 1970] gives an absolute scintillation efficiency for electron excitation of 1.3 per cent, which is equivalent to 230 ± 20 eV of ionising energy per photon (units used by Spowart). From data more recently reported for an NE905 glass [Jensen and Czirr 1983], the scintillation efficiency for electron excitation was estimated to be 1.5 per cent (202 ± 30 eV per photon). The present result is in good agreement with both values.

The scintillation responses to thermal neutrons of glasses containing the range of lithium concentrations used here have been reported by Spowart [1976]. Estimates of the intrinsic resolutions obtained from these data by the author are listed in **table 3**. Although there are significant differences between these estimates and the present values for individual glasses, a similar correlation was observed between $\eta^2(I)$ and lithium concentration; also noted was the absence of any correlation with isotopic concentration.

The mechanism responsible for the scintillations in the glass is dependent on the atomic levels within its microcrystalline structure [Spowart 1976]. Hence significant changes in scintillation response should be expected for the radical differences in the glass composition associated with the different lithium concentrations (**table 1**). The trend observed in the present investigation conforms with that predicted by the theory. However, no differences in response should be expected for glasses of different lithium enrichment but having the same chemical composition. Hence the variations in S and $\eta(I)$ observed in the present investigation, and the differences in $\eta(I)$ observed between the present data and those of Spowart [1976], indicate that there are significant microscopic variations in the internal structure of glasses with the same nominal macroscopic composition.

5. CONCLUSIONS

A feasibility study has been conducted at Lucas Heights on the use of lithium glass scintillators for the detection of tritium production in experimental fusion blanket assemblies. The scintillation response characteristics of the lithium glasses to electron excitation were measured to provide experimental and theoretical data which can be utilised for the compensation of the background effects produced by the neutron-gamma fields in fusion blanket assemblies. The characteristics of importance are the scintillation efficiency and the intrinsic resolution of the glass.

At a specific location within the fusion blanket (*i.e.* in given energy distributions of neutrons and gammas), the profile of the background signals depends not only on the chemical composition of the glass but also on structural differences which can arise from local inhomogeneities introduced during manufacture. The magnitude and resolution of these signals determines the extent of their overlap with those produced by the charged particles released in the ^{6}Li (n,α) T and ^{7} Li(n,n′α) T reactions [Dalton 1987].

For background compensation methods based on measuring the differences between the responses of glasses enriched in either ^{6}Li or ^{7}Li [Yamaguchi and Nakumura 1986] in the mixed neutron/gamma field of a fusion blanket assembly, it is essential that the different enrichments should be selected from glasses which have the same chemical composition (same nominal lithium concentration). This ensures that the same range of interactions occur in the two glasses.

To minimise response variations arising from structural differences in glasses of the same chemical composition, the glasses must be matched on the basis of their observed responses. In the present investigation, the pair of glasses containing the lowest concentration of lithium — NE902 and NE903 — had the lowest variation in both scintillation efficiency and intrinsic resolution with enrichment. These glasses will therefore be used in future tritium production measurements.

6. ACKNOWLEDGEMENTS

My thanks are due to R. J. Blevins for valuable assistance in setting up the detection system and the measurement of the glass responses, and to N. Chankow (on attachment from Chulalongkorn University, Thailand) for assistance in the analysis of the measured data.

7. REFERENCES

Bisi, A., Sappa, L. [1958] - Statistical spread in pulse size of scintillation spectrometer. *Nucl. Instrum. Methods,* 3: 17-24.

Breitenberger, E. [1955] - Scintillation spectrometer statistics. In *Progress in Nuclear Physics* (ed. Frisch, O.), Pergamon Press, London, New York.

Case, K.M., De Hoffmann, F., Placzek, G. [1953] - Introduction to the Theory of Neutron Diffusion, Vol 1. Los Alamos Scientific Laboratory, Los Alamos, New Mexico.

Dalton, A.W. [1987] - Measurement of the responses of small lithium glass scintillators to protons, deuterons and alpha particles. AAEC/E641.

Davisson, C.M. [1965] - Interaction of gamma-radiation with matter. In *Alpha, Beta and Gamma-ray Spectroscopy,* Vol. 1 (ed. Siegbahn, K.), North Holland Publishing Co., Amsterdam, pp. 37-78.

Evans, R.D. [1958] - Compton effect. In *Handbuch der Physik,* Vol 34 (ed. Flugge, S.) Springer-Verlag, Berlin, pp. 218-298.

Flynn, L.E., Glendenin, L.E., Steinbert, E.P., Wright, P. M. [1964] - Scintillation detection of gamma radiation. *Nucl. Instrum. Methods,* 27:13-19

Garlick, G.F., Wright, G.T. [1952] - Characteristics of scintillation counters. *Proc. Phys. Soc.*, B65: 414-421.

Jensen, G. L., Czirr, J.B. [1983] - Gamma-ray sensitivity of ^{6}Li-glass scintillators. *Nucl. Instrum. Methods,* 205:461-463.

Katz, L, Penfold, A.S. [1952] - Range-energy relations for electrons and the determination of beta-ray end point energies by absorption. *Rev. Mod. Phys.*, 24:29-44.

Prescott, J.R. [1963] - Photomultiplier single-electron statistics and the shape of the ideal scintillation line. *Nucl. Instrum. Methods,* 22:256- 268.

Spowart, A.R. [1969] - Measurement of the absolute scintillation efficiency of granulated and glass neutron scintillators. *Nucl. Instrum. Methods,* 75:35-42.

Spowart, A.R. [1970] - Measurement of the gamma sensitivity of granulated and glass neutron scintillators and films. *Nucl. Instrum. Methods,* 82:1-6.

Spowart, A.R. [1976] - Neutron scintillation glasses: part 1. Activation by external charged particles and thermal neutrons. *Nucl. Instrum. Methods,* 135:441-453.

Whittlestone, S., [1979] - The Energy Spectrum of Neutrons in a Pulsed Fast Assembly. Ph.D Thesis, University of Wollongong.

Yamaguchi, S., Nakamura, T. [1986] - *Proc. 7th Topical Meeting on the Technology of Fusion Energy,* Reno, Nevada, June 15-19.

TABLE 1
DETAILS OF LITHIUM GLASS SCINTILLATORS

Glass	Total Lithium (wt %)	^{7}Li/Li (wt %)	Nominal Concentration of Materials (wt %)				
			SiO_2	MgO	Al_2O_3	Ce_2O_3	Li_2O
NE901	2.4	7.50	56	24	11	4	5
NE902		5.00					
NE903		99.99					
NE904	6.6	7.50	60	6	18	4	14
NE905		5.00					
NE906		99.99					
NE907	7.5	7.50	79	-	-	4.6	16
NE908		500					
NE909		99.99					
NE912	7.8	5.00	77	-	-	4.6	18
NE913	8.3	99.99					

TABLE 2
DEPENDENCE OF RESPONSE ON GLASS THICKNESS AND GAMMA ENERGY

Glass Thickness (mm)	Mean Chord Length (mm)	Maximum Electron Energy (MeV)	Gamma Energy (MeV)			
			0.5	1.0	2.0	10.0
			Detection Efficiency (%)			
1.5	2.43	0.91	4.9	3.8	2.7	1.1
3.0	4.07	1.23	8.1	6.2	4.4	1.8
6.0	6.17	1.57	12.0	9.3	6.6	2.7
Compton Edge Energy (MeV)			0.3	0.8	1.8	9.8

TABLE 3
SCINTILLATION RESPONSES OF LITHIUM GLASSES TO ELECTRON EXCITATION

Glass Type	Scintillation (eV/photon)	Efficiency (%)	Intrinsic Resolution Dalton (%)	Intrinsic Resolution Spowart [1976] (%)
NE901	162	1.94	14.3	11.7
NE902	185	1.70	14.6	13.1
NE903	184	1.71	14.9	
NE904	180	1.74	15.4	13.5
NE905	231	1.36	19.0	19.0
NE906	244	1.29	19.9	
NE907	297	1.06	18.3	15.7
NE908	384	0.82	16.0	20.2
NE909	325	0.95	19.2	21.5
NE912	344	0.91	21.6	23.6
NE913	430	0.73	23.0	30.1

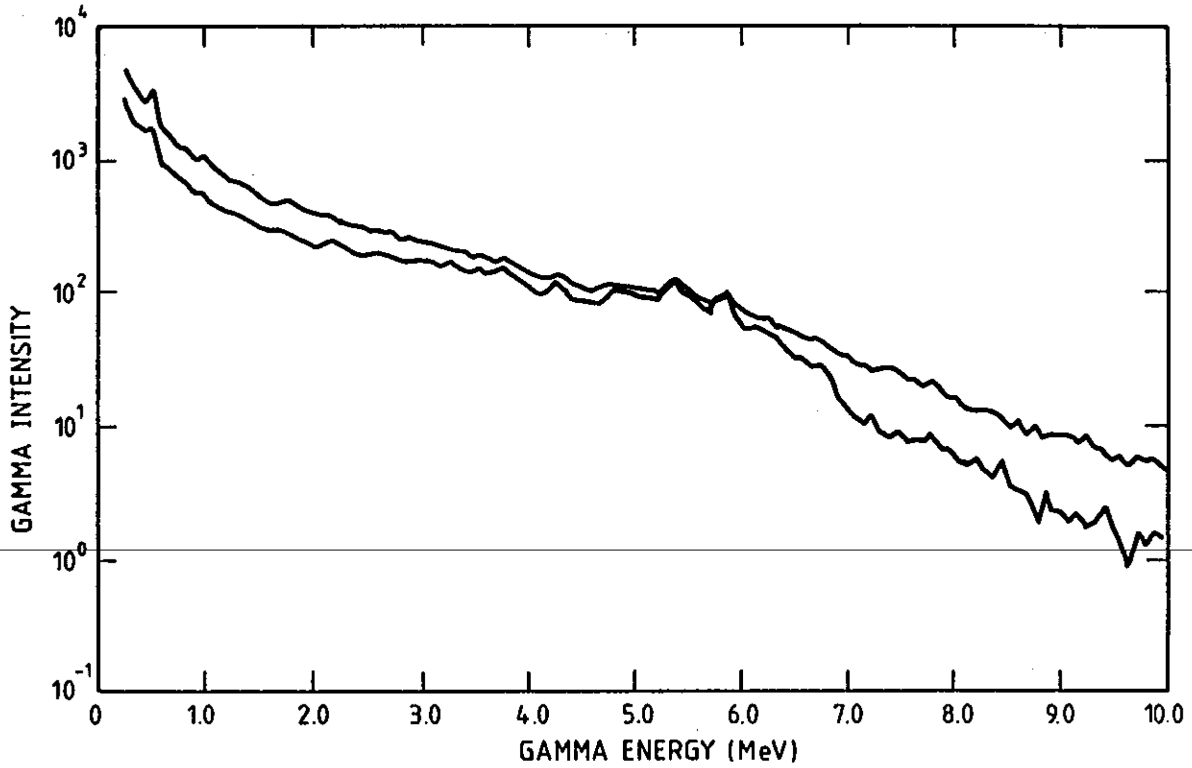

Figure 1 Energy distribution of gamma radiation in a fusion breeder blanket

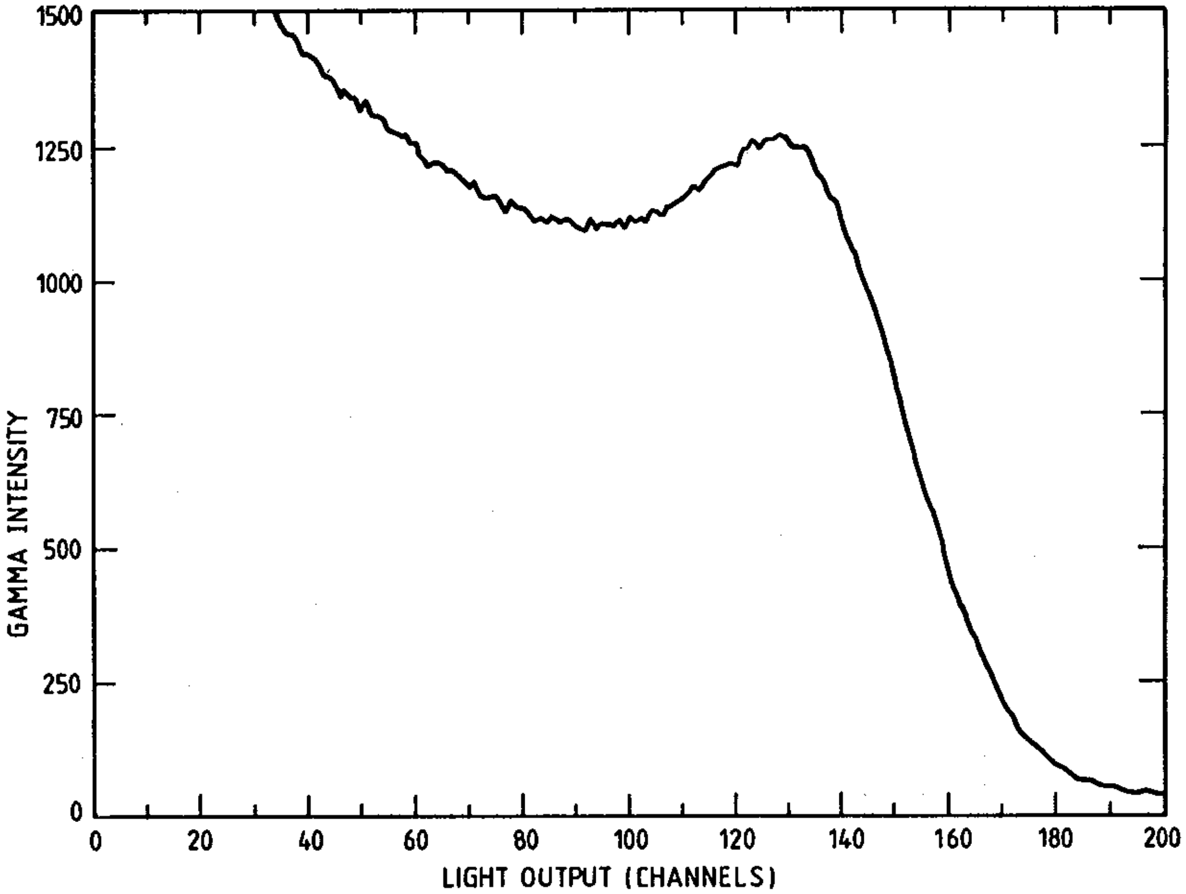

Figure 2 Scintillation response of the NE902 glass to a ^{137}Cs source

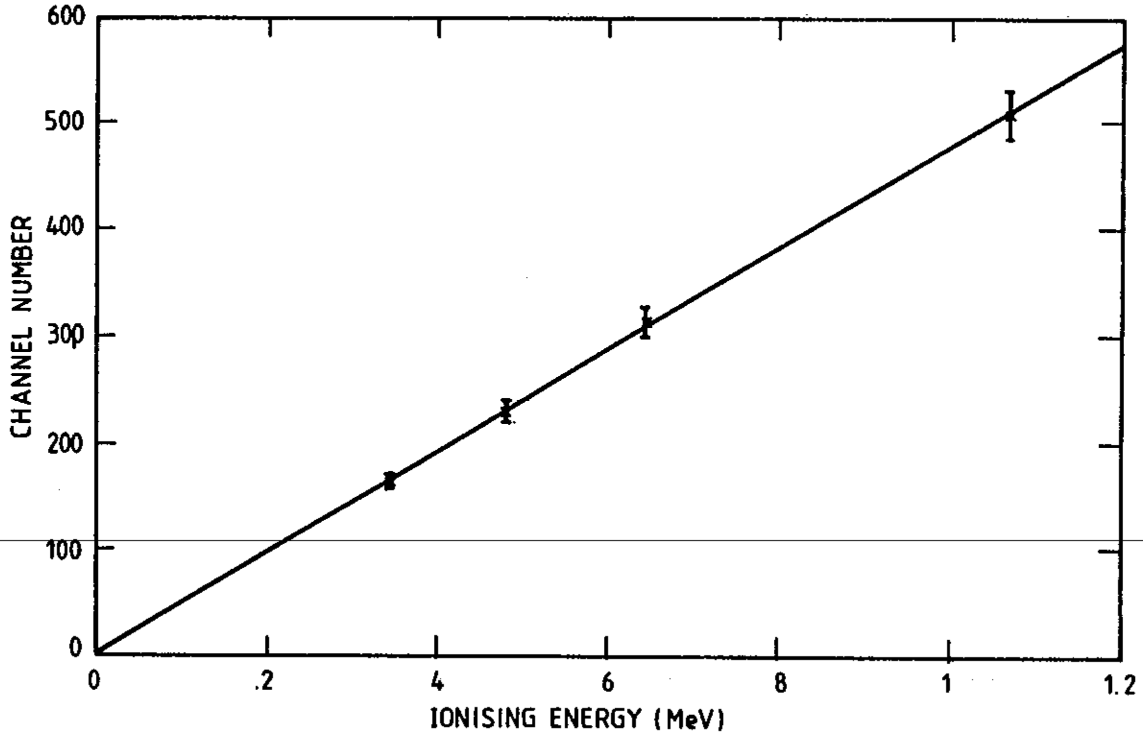

Figure 3 Variation of light output with electron energy (NE902 glass)

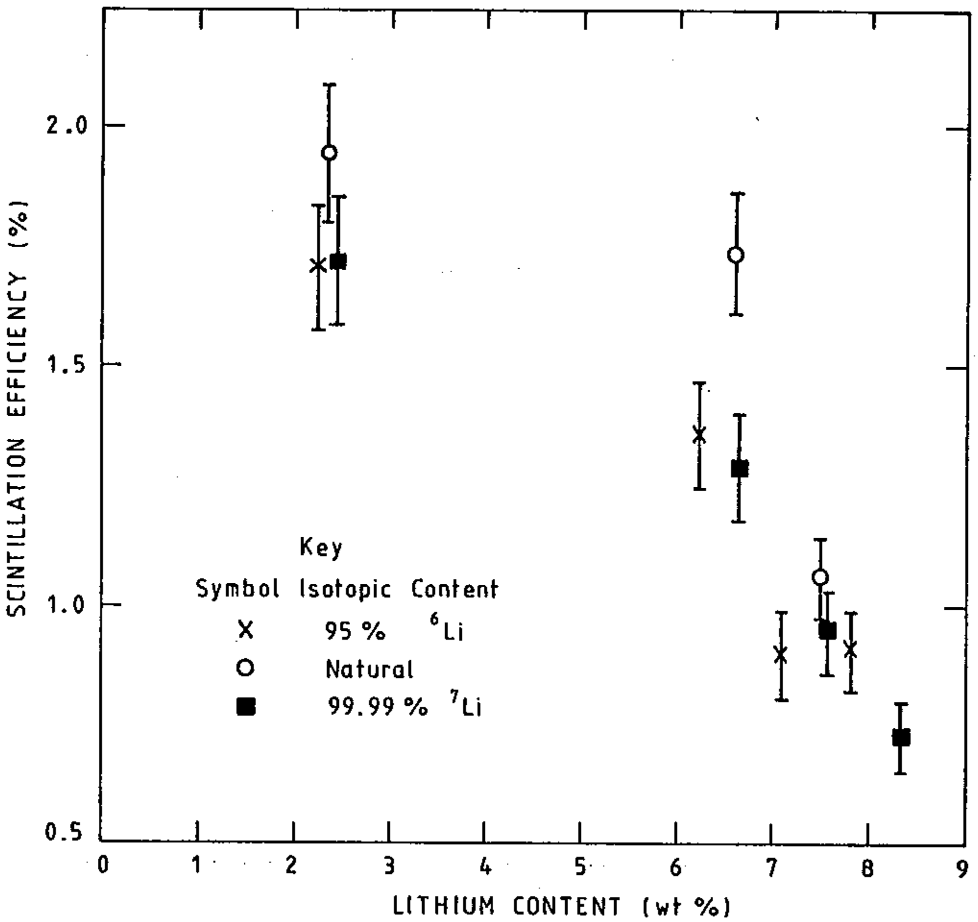

Figure 4 Variation of scintillation efficiency with lithium content