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Preprint
RAL-P-95-003

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October 1995

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ISSN 1358-6254

DENSE, FAST, RADIATION-TOLERANT FLUORO-HAFNATE GLASS SCINTILLATORS FOR ELECTROMAGNETIC CALORIMETERS IN HIGH ENERGY PHYSICS*

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ABSTRACT

Over 200 different compositions of heavy metal fluoride glasses have been produced and evaluated as possible candidates for the active medium in homogeneous electromagnetic calorimeters for high energy particles. Promising glasses, based on mixtures containing hafnium fluoride, have densities of 6 g cm^{-3}, radiation lengths of 1.6 cm, and good optical transmission from the UV to the IR. Glasses containing the Ce^{3+} ion are fast scintillators, with properties similar to those of pure cerium fluoride crystal. We have made glasses which have up to 14% of the light yield of CeF_3, fast scintillation characterised by typical exponential decay constants of 10 ns and 25 ns, and a temperature dependence of light yield of -0.4%/°C over the range -10°C to 60°C. We have carried out a detailed investigation of the effects of gamma irradiation (up to doses of 6 kGy) on the optical transmission, and systematic studies into the role of indium and other elements on radiation tolerance and light yield. Some glasses have shown complete optical annealing of radiation-induced absorbance when exposed to UV light.

INTRODUCTION

Almost all current and planned experiments in particle physics incorporate detectors for measuring the directions and energies of high energy electrons and photons - so called electromagnetic calorimeters (often abbreviated to ECAL). The technologies employed in these detectors fall into one of two categories; 'sampling' and 'homogeneous'. In sampling detectors many layers of an active medium (plastic scintillator, gas or liquid ionisation device, semiconductor diode) are interleaved with a dense material having high atomic number, such as lead or tungsten. Homogeneous detectors have been constructed using silicate glass loaded with barium or lead; large, dense scintillating monocrystals of materials such as sodium iodide, caesium iodide or bismuth germanate; or a dense liquified noble gas such as krypton. As a rule of thumb, homogenous detectors give better energy resolution but are more expensive when compared with their sampling counterparts. Furthermore, among the available homogenous media, heavy silicate glasses are cheaper than crystals but give somewhat inferior energy resolution.

The work described here was motivated by the desire to develop a new homogenous medium for high energy electromagnetic calorimetry, which would give an energy resolution approaching that of crystals, at a significantly lower cost. The particular application in mind was the CMS (Compact Muon Solenoid) experiment at the LHC (Large Hadron Collider) recently approved for construction at CERN. The calorimeter for such an experiment must be fast (signals of 25 ns duration or less), dense (a radiation length of 2 cm or less), radiation hard (able to survive a dose of 10 kGy) and realisable on a large scale (tens of cubic metres of detector are required). The most promising materials we have investigated are glasses based on the hafnium fluoride matrix.

FLUORIDE GLASSES

Heavy metal fluoride glasses were discovered in 1974 [1] in the course of investigations of crystalline laser hosts. Many of these glasses have very good optical transparency, and interest focused on their potential for use in extremely low loss optical fibres for communications. A formulation which has

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*Paper presented at the International Conference on Inorganic Scintillators and Their Applications, Delft 1995 been extensively studied is ZBLAN (ZrF_4, BaF_2, LaF_3, AlF_3, NaF) which is relatively stable against crystallisation during production, allowing kilogram scale blocks to be cast.

Fluoride glasses readily accept transition metals and lanthanides into the matrix, offering the possibility that they might be made to scintillate by doping with a fluorescent ion such as Ce^{3+}. ZBLAN has a radiation length of 3.2 cm which is too long for applications at LHC. Nevertheless, it might have proved of interest for other particle physics experiments, however, our early work showed that replacing lanthanum with cerium did not result in a measurable scintillation yield from this glass [2]. Mixtures based on HfF_4 rather than ZrF_4 also form stable glasses and these have much shorter radiation lengths (typically 1.6 cm). Furthermore, in 1992 Devitsin et al [3] showed that doping with CeF_3 at the level of a few molar percent produces a significant scintillation yield and this has led us to concentrate our investigations on these materials.

GLASS PRODUCTION

Glass samples were prepared at Sheffield University from their component metal fluorides, which were in the form of small crystals or powders (HfF_4 was obtained from Johnson Matthey [4], Merck [5] and Moscow [6], all other metal fluorides were supplied by Merck). Typically, 10 g batches were prepared giving 20 x 10 x 6 mm^3 finished samples. The glass production techniques are described in greater detail elsewhere in these proceedings [7].

LIGHT YIELD

The scintillation yields were measured relative to a CeF_3 crystal using a secondary charged-particle beam at the proton synchrotron accelerator of the ISIS neutron spallation source at the Rutherford Appleton Laboratory. The beam line was tuned to accept protons with a mean momentum selected in the range from 540 to 750 MeV/c. At these momenta the protons are below threshold for directly producing Cherenkov light in the sample (the threshold is 840 MeV/c for a refractive index of 1.5). This is an important consideration since even a small contribution from Cherenkov photons can bias fits to scintillation decay curves. Scintillation light was detected by a Thorn EMI 9814QKB photomultiplier. The photomultiplier signal was recorded on a 1 G Sample/s digital oscilloscope, using a gate width of 500 ns.

For a given basic glass composition, the light yield was found to increase monotonically with cerium content up to the maximum concentration that could be incorporated into a stable glass. Typically, a glass containing cerium fluoride at a concentration of 5% (molar) gives a light output per MeV of

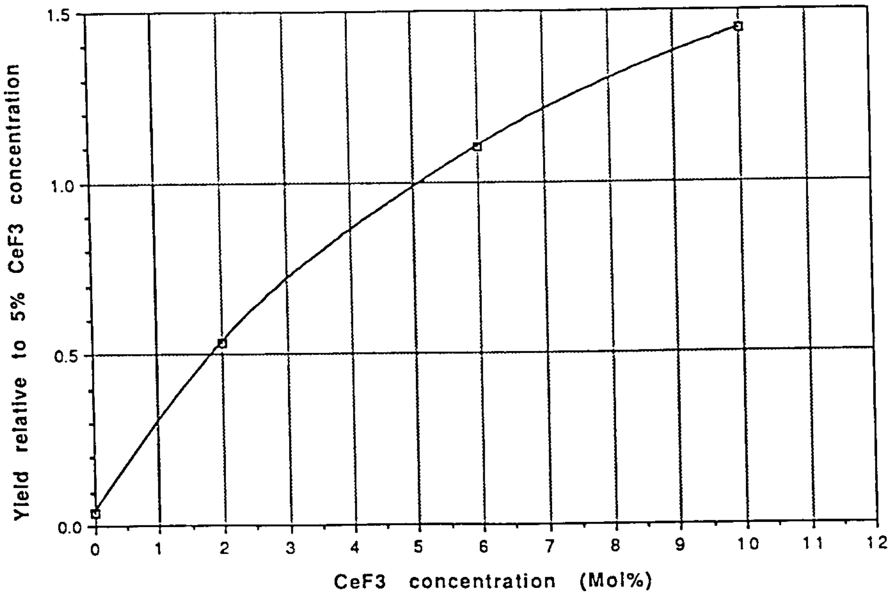

Figure 1 The measured light yield plotted against CeF_3 concentration, for a set of glasses of composition 'HBCeAN'. The yield has been normalised to unity for 5% CeF_3 concentration.

energy deposited which is approximately 10% that of a cerium fluoride crystal. Figure 1 shows the variation of light yield with cerium fluoride concentration for a set of glasses with a composition HBCeAN.

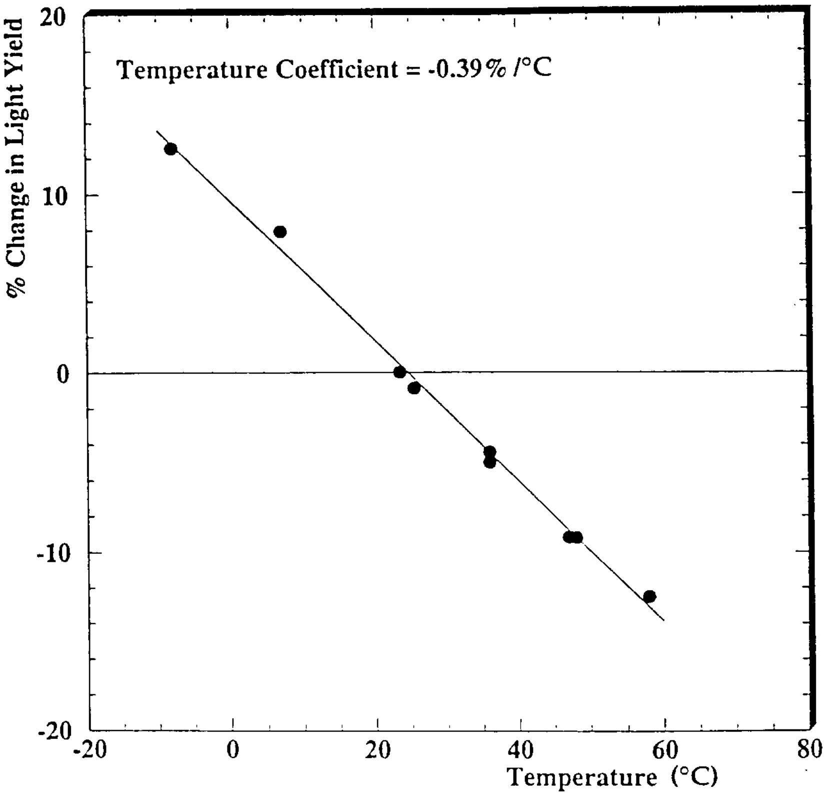

Figure 2 The measured variation of light yield with temperature for a fluorohafnate glass doped with CeF_3.

With the aid of a Peltier cooling/heating device, a set of measurements was made for one of the samples (HBCeA), at temperatures in the interval from -10 to + 60°C. The light yield was found to decrease linearly with increasing temperature over this range by approximately -0.4%/°C (figure 2).

DECAY TIME SPECTRA

Scintillation decay time spectra were obtained by two methods. In one approach, several thousand individual digitised wave forms, recorded with protons at ISIS, were summed to give an average pulse shape. The other measurement, performed at Brunel University, used a standard delayed single-photon technique, with an additional multi-photon veto logic. The sample was excited by an annihilation photon from a 10 μCi Na^{22} positron source, the other photon was detected in a BaF_2 crystal which started a fast time-to-amplitude converter. The data were collected over a time window of 2000 ns, with a single-photon timing resolution of 1.40 ns.

Figure 3 shows a typical decay time distribution obtained with protons. In the range from 0 to 250 ns, the curve is well fitted by the sum of three exponentials:

$$I(t) = I_0\left(ae^{-t/\tau_1} + be^{-t/\tau_2} + ce^{-t/\tau_3}\right)$$

where $\tau_1 = 11$ ns, $\tau_2 = 25$ ns, $\tau_3 = 62$ ns and the coefficients a, b and c are in the ratio 0.16:1.0:0.26.

Since the three terms are strongly correlated in the fit, the decay spectrum is more usefully characterised by considering the fraction of the light yield contained within a given time interval. Integrating under the curve, one finds that 53% of the light is emitted in the first 25 ns. This result is very similar to that obtained with a single crystal of CeF_3.

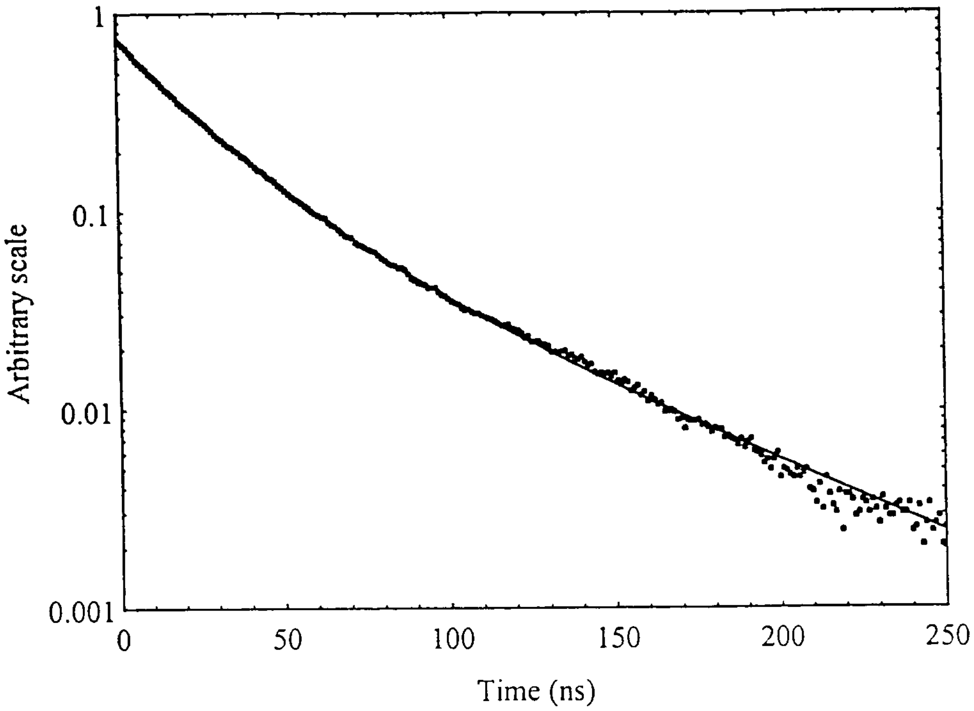

Figure 3 The measured decay time spectrum for scintillation light from a sample of Ce doped fluorohafnate glass (HBCeALi). The smooth curve is a fit to the data and is the sum of three exponential terms: $I(t) = I_0(ae^{-t/\tau_1} + be^{-t/\tau_2} + ce^{-t/\tau_3})$ where $\tau_1 = 11$ ns, $\tau_2 = 25$ ns, $\tau_3 = 62$ ns and the coefficients a, b and c are in the ratio 0.16:1.0:0.26.

TRANSMISSION AND EMISSION SPECTRA

Emission spectra have been measured using X-ray (~ 9 keV) and ultraviolet light (284 nm) excitation, with similar results. Figure 4 shows the fluorescence spectrum obtained for a fluorohafnate glass (HBCeA) containing CeF_3 at a concentration of 8% (molar). The spectrum is dominated by an emission band centred at 320 nm, which is attributed to radiative transitions from the lowest excited 5d-level of Ce^{3+}, to the 4f ground state.

The optical transmission as a function of wavelength is shown on the same plot. It can be seen that the emission spectrum lies close to the band-edge. Good optical transmission just above the cut-off is therefore crucially important for detectors using large blocks of glass, if significant self absorption of scintillation light is to be avoided.

RADIATION DAMAGE

Radiation damage in glasses is characterised by the development of optical absorption bands which result from the creation and population of defects known as colour centres. The development of these absorption bands is consistent with the irradiation creating and populating new defect sites and the sites relaxing either spontaneously or as a result of irradiation, or both. The absorbance at a particular wavelength saturates as a function of dose.

In an LHC experiment the radiation dose in the electromagnetic calorimeter will be dominated by electromagnetic showers produced by high energy photons from π^0 decays. As a result, the dose will vary with depth in the calorimeter (being greatest at the position of the average shower maximum) and will be higher close to the direction of the colliding beams (the 'forward' regions) than at larger angles with respect to this axis (the 'central' region). The estimated dose in the central region, averaged over the first eight radiation lengths of the detector, is 0.4 kGy/year. In order to investigate the ability of our glasses to withstand these large radiation levels, samples were irradiated with a 4 Ci Co^{60} source at Brunel. Doses of up to 6 kGy were delivered in 5 days (at a dose rate of 12 m Gy/s), corresponding to more than 10 years of operation in the central region at LHC.

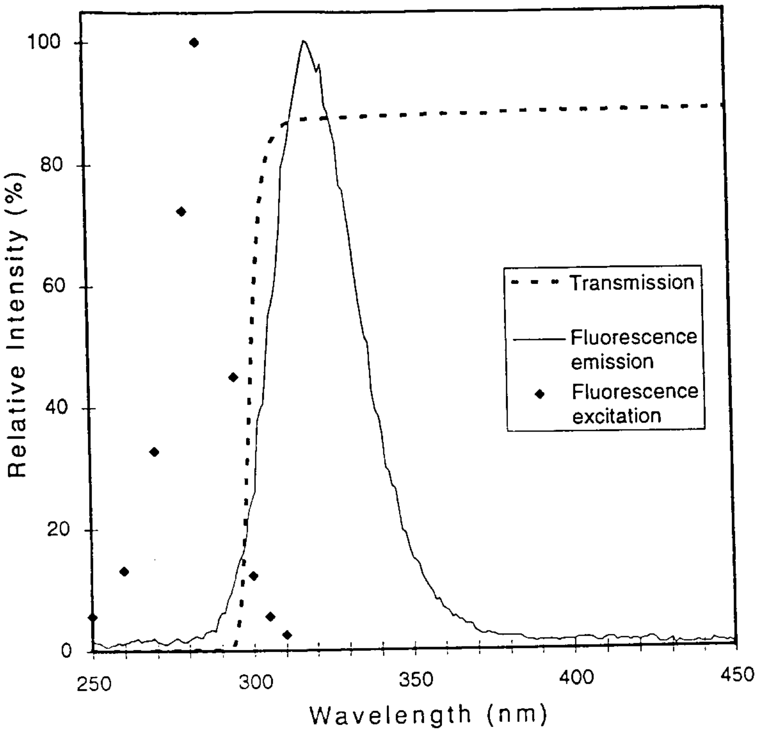

Figure 4 Fluorescence excitation and emission spectra for a cerium doped fluorohafnate glass together with the optical transmission measured for the same sample.

All undoped fluorohafnate glasses studied proved to be quite susceptible to radiation damage, the internal transmission at 325 nm of a 5 mm thick sample typically falling to 5% of its initial value after 6 kGy. Studies were therefore made to determine the effect of various dopants (Fe, Ga, Nb, In, Pr, Nd, Sm, Eu, Gd, Ho, Er, Yb, Lu) on radiation hardness. In some cases the effect was quite dramatic. Thus for example adding 1.5% (molar) of EuF_3 to HBLA resulted in a glass which exhibited negligible damage at wavelengths above 400 nm for doses in excess of 6 kGy. Unfortunately strong damage was still observed in the Ce^{3+} emission region from 300 to 350 nm.

The dopant which was found to minimise radiation damage in the Ce emission region is indium, the magnitude of the effect depending quite sensitively on the concentration (figure 5). Curve II in this figure shows how the radiation-induced optical attenuation (at 350 nm), measured immediately after a dose of 2.4 kGy (Co^{60} γ-ray), varies with indium concentration. The addition of indium has the undesired effect of reducing the light yield from Ce doped glass, as can be seen from curve I. However, for an indium concentration of 0.5%, which maximises the radiation hardness, the loss in light yield (from a small sample) is only 30%.

Since the radiation dose received by a calorimeter at LHC would be spread over the ten year lifetime of the experiment, the effect of thermal annealing over a long interval at room temperature should also be taken into account. Curve III in figure 5 shows how the radiation-induced attenuation length varies with indium concentration for a much larger dose (7 kGy delivered in 7 days) followed by storage for 4.5 months in the dark at room temperatures. Comparing curves II and III it can be seen that self annealing largely offsets the effect of the bigger radiation dose for indium concentrations of 0.8% and above. At lower indium concentrations self annealing occurs to a greater degree and this is particularly evident for the point at 0.2% concentration. However, the optimum indium concentration still appears to be close to 0.5%, even allowing for self annealing.

The above studies were performed using small samples of glass (typically 1 cm^3). In order to obtain a better understanding of how the results would translate to the performance of a glass calorimeter, the effect of irradiation on light yield was measured using a much larger block of glass (HBCeALi doped with 0.5% InF_3). The block was 13 cm (8.2 radiation lengths) long with a rectangular cross section (3 x 1 cm^2). For the light yield measurements it was wrapped in Tyvek and a 2" phototube was

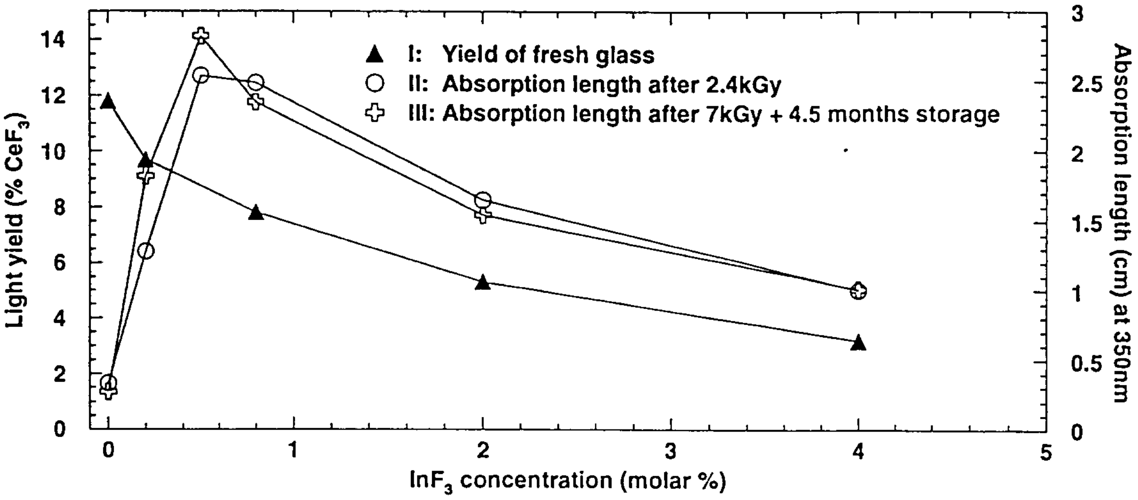

Figure 5 The effect of InF_3 doping on the light yield and radiation hardness of a glass with basic composition 'HBCeAN', containing 5% (molar) CeF_3.
Curve I (triangles) shows the variation of light yield versus InF_3 concentration. The yield is normalised to the yield obtained from a crystal of CeF_3 of similar dimensions to the glass samples.
Curve II (open circles) shows the variation of radiation-induced optical attenuation at 350 nm, measured immediately after a dose of 2.4 kGy. (Co^{60}-γ).
Curve III (crosses) shows the optical attenuation induced by a dose of 7 kGy, followed by 4.5 months storage in the dark at room temperature.

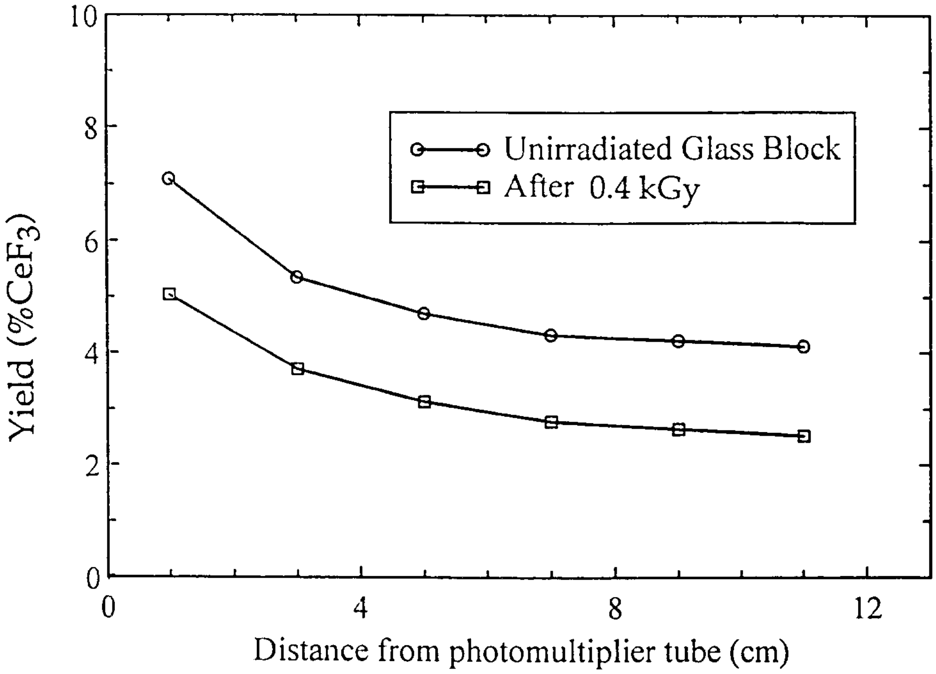

Figure 6 The light yield as a function of position along the length of a block (dimensions 13 x 3 x 1 cm^3) of fluorohafnate glass (HBCeALi) doped with 0.5% (molar) InF_3.
The upper curve was obtained before irradiation. The lower curve was measured immediately following a 0.4 kGy dose (Co^{60}-γ) delivered approximately uniformly over the volume of the block.

mounted on one end. The proton beam was directed through the 1 cm dimension at several points along the length. The block was then irradiated with a dose of 0.4 kGy in 40 hrs (~ 1 year dose at LHC), delivered approximately uniformly over the whole volume, and the light yield remeasured.

The results are shown in figure 6. Both before and after irradiation the light yield shows a strong variation as a function of distance from the phototube in the region close to the tube, whereas further from the tube the yield varies rather slowly. The loss of light from the far end following irradiation is 35%. A full sized detector would be 3 times longer than the block tested, however, the radiation dose at LHC falls off rapidly after the first 8 radiation lengths. On the basis of these data we conclude that this particular glass would satisfactorily survive one year of operation in the central rapidity region at LHC. However, ten years of operation would require some method of *in situ* annealing to be devised.

OPTICAL ANNEALING

All the glasses were stored and irradiated in complete darkness to avoid any possibility of photo-induced bleaching. After spontaneous recovery of absorbance had stabilised, several glasses were illuminated with light from a 150 W Hg arc lamp. Interference filters were used to select the 365 nm, 435 nm and 577 nm lines in the Hg spectrum. Lines above 400 nm were observed to cause little measurable change in absorbance, but the 365 nm line was extremely effective in reversing the radiation induced optical damage in some fluoride glasses. Some glasses were found to recover fully (figure 7); whereas others, including those doped with indium, recovered only partially.

Complete optical annealing of radiation damage in a scintillating fluoride glass has previously been reported by the Crystal Clear Collaboration[8].

PROTOTYPE GLASS DETECTOR

A composite block of glass (13 x 3 x 3 cm^3), fabricated from three plates similar to that used for the irradiation study, has been tested in a high energy electron beam at CERN. The beam was directed on to the centre of the front face and the light was detected with a 2" phototube (9814QKB) mounted on the rear face. The measured response to 10 and 50 GeV electrons was found to be in good agreement with that predicted by a GEANT [9] simulation of the energy deposition in this detector (figure 8).

CONCLUSIONS

We have demonstrated that cerium doped fluorohafnate glass could be used as the active medium in a high performance electromagnetic calorimeter for many particle physics applications. Radiation damage would be a limitation to use in the harsh environment at LHC although this might be overcome by optical annealing *in situ*.

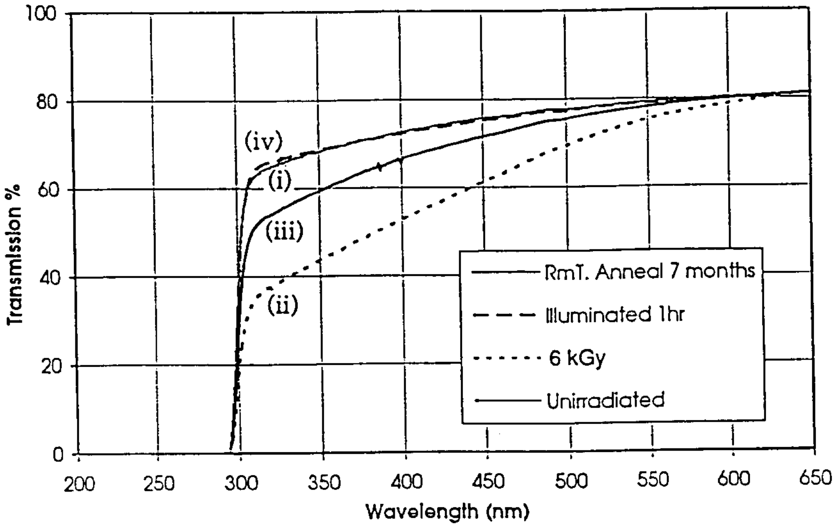

Figure 7 The measured external transmission of a fluoro-hafnate glass; (i) unirradiated, (ii) after 6 kGy, (iii) after annealing in the dark at room temperature for 7 months (where strong recovery is observed), and (iv) after illuminating with a mercury arc lamp for one hour. The glass has fully recovered after one hour of optical annealing.

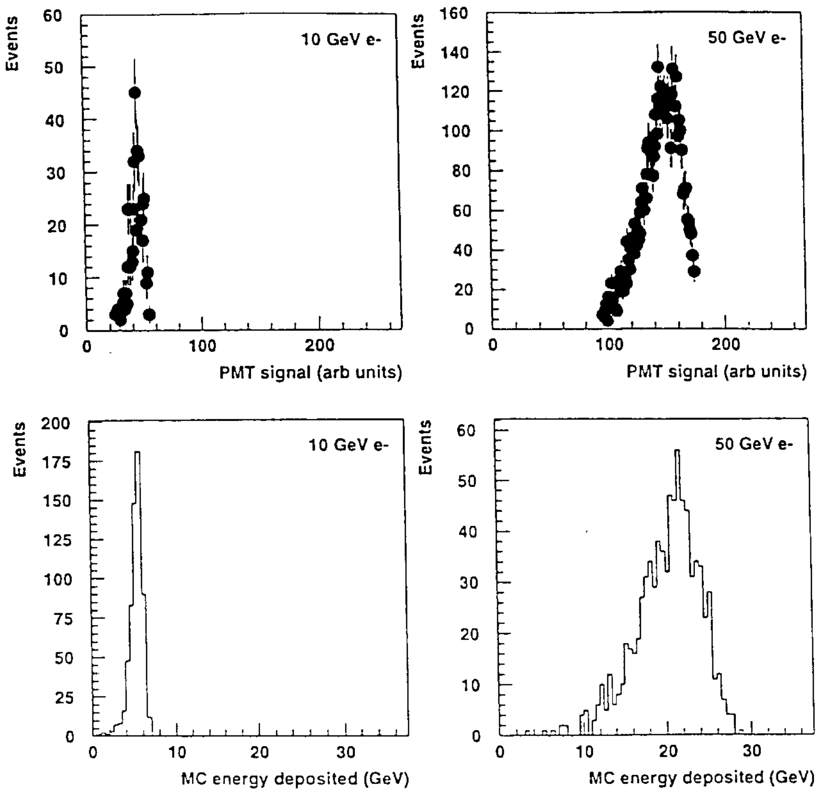

Figure 8 The measured (upper) and Monte Carlo simulated (lower) response of a composite glass block of CeF_3-doped fluorohafnate glass to 10 GeV and 50 GeV electrons. The block was 3 x 3 cm^2 in section and 13 cm ($8.2X_0$) long.

ACKNOWLEDGEMENTS

We thank E G Devitsin, V A Kozlov, V N Makhov and S U Potashov from the Lebedev Institute and L N Dmitruk from the General Physics Institute in Moscow for many helpful discussions, for supplying some of the HfF_4 raw material and for suggesting the glass formulation used for the CERN beam tests. We are grateful to S Sheikh from the Department of Physics at Brunel University, for measuring the fluorescence spectra of the HBCeA glass and to B W Kennedy for analysing data recorded at ISIS.

This work has been supported by the UK Particle Physics and Astronomy Research Council.

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