# HIGH AND LOW DOSE RATE IRRADIATIONS OF SCINTILLATORS AND WAVE LENGTH SHIFTERS

A.Dannemann, U.Holm,T.Marckmann,K.Wick
I.Inst.f.Experimentalphysik, Universität Hamburg,
Luruper Chaussee 149, D-2000 Hamburg 50 and
M.Rohde
Deutsches Elektronen-Synchrotron DESY,
Notkestr. 85, D-2000 Hamburg 52

## 1 Introduction

The calorimeter of the ZEUS detector[1] at the HERA collider consists of depleted uranium (DU) plates interleaved with plastic scintillators (SCI) SCSN-38[2]. The SCI are read out via wavelength shifters (WLS) Y-7 in PMMA with a UV absorber[2]. Both plastic materials have to withstand a radiation background from the collider, especially hadrons hitting absorbers, which has been estimated for stable machine operating conditions to about 100 Gy/a. Additionally they are exposed to the low dose rate of several ten Gy/year from the DU. Both the yield of the scintillation light and also the transmission through the material may decrease with an increasing dose and so lead to a degraded calorimeter resolution and homogeneity. Even the low DU radiation dose may strongly affect the quality of the optical components[3]. Therefore various investigations concerning the radiation stability of SCI and WLS have been carried out. The SCI have been irradiated with $^{60}Co$-sources, a 25 MeV proton beam and, for very low dose rate investigations, plates of DU. The applied doses were 1, 10 and 24 kGy, the latter with dose rates between 30 and 1000 Gy/h. The DU radiation dose was 42 Gy with 1.6 mGy/h. The samples have been irradiated and stored under different atmospheres(e.g. air, oxygen, nitrogen, argon).

Diagnostic tools for the change in absorption length and loss in light yield were a spectrophotometer, and a movable UV lamp, witch can excite the different fluors seperately.

## 2 Short-term irradiations

The results of the short-term high dose rate proton and $^{60}Co$ irradiations show a very similar behaviour for quite different materials. The radiation induces additional absorption centers (free radicals) which shorten the absorbtion length of the scintillation light. No influence of the surrounding gas atmosphere on the type and concentration of the radicals was seen for dry air, nitrogen and argon[4]. Under an inert gas (Ar, $N_2$, $CO_2$) atmosphere after irradiation, one has a slow decrease of the density of the induced absorption centers (recovery) in the SCI SCSN-38, while for the WLS Y-7 and K-27 in PMMA no recovery at all is observed. In the presence of oxygen the free radicals form peroxide radicals which do not absorb the emitted light and one generally has a strong recovery of SCI and WLS to a permanent damage which is much smaller than the initial one. The recovery time constant strongly depends on the oxygen pressure, the thickness of the sample and the base material(diffusion constant, radical concentration) and is quite different for the polystyrene based SCSN-38 ($\sim$ 1 day, 2mm thick) and the acrylic WLS ($>$ 1 year, 2 mm thick). This can be well understood by a simple oxygen diffusion model [11]: the diffusion constant of polystyrene is about a factor ten higher whereas the created radicals per dose are 60 times more for PMMA (see chapter 5). Another radiation effect is the destruction of the dye molecules which leads to a reduced fluorescence efficiency.

The radiation induced additional absorption coefficient $\Delta\mu$ , measured with a xenon lamp device, can be described by a sum of a time dependent term (recovery) and a constant term which describes the permanent damage. Both are proportional to the applied dose D (at least for our doses):

---

This work was supported by the German Federal Minister for Research and Technology (BMFT) under the contract number 05 5 HH 17I.

$$\Delta\mu(D,t) = a \cdot D \cdot f(t) + b \cdot D, \qquad (1)$$

where t is the time after the end of the short-term irradiation. The function $f(t)$ with $f(t = 0) = 1$ and $f(t = \infty) = 0$ describes the decay of the initial damage by diffusion of oxygen. The change $\Delta\mu(\lambda)$ of the spectral absorption coefficient , measured with a spectrophotometer, behaves in the same way.

Table 1 shows a summary of the high dose rate low term irradiations of SCSN-38, Y-7 and K-27. Irradiations with $^{60}Co$ and 25 MeV protons and measurements with the xenon lamp and the photospectrometer are compared. The results are compatible especially if one takes into account that the induced additional absorption coefficient $\Delta\mu$, measured with the xenon lamp, depends on thedifferent dimensions of the sample and on the fits to the attenuation curves, which also had been different.

For the ZEUS calorimeter follows that 10 years after operation in air the attenuation length of the SCI will change for about 5% which is unnoticeable for the only 20 cm long SCI. For the WLS Y-7 one expects in the direct vicinity of the beam pipe a loss of light transmission of $< 17\%$ for the longest (2 m) WLS.

The loss in SCSN-38 fluorescence light yield due to destroyed fluors is $(5 - 9) \times 10^{-4}\%$ Gy$^{-1}$ for irradiation in dry air, measured with different methods and SCI thicknesses. In argon the loss is only half as big [12].

Table 1: *Summary of the high dose rate irradiations (see eq. 1 and 2). The applied doses were 24 kGy for the $^{60}Co$ exposure with rates of 1 - 10 kGy/h and 10 kGy with rates of 2 - 10 kGy/h for 25 MeV protons. The irradiation and storage has been performed in (dry) air. The fluor concentration of the WLS was 30 ppm Y-7 in PMMA and 100 mg/l K-27 in PMMA - except for the p irradiations where we had 47 ppm or 150 mg/l. The spectral absorption coefficient $\Delta\mu(\lambda)$ is shown for $\lambda = 430$ nm (SCSN-38) and 510 nm (WLS). All quantities are given in $10^{-6}cm^{-1}Gy^{-1}$*

| | | $^{60}Co$ photospect. | $^{60}Co$ xenonl. | 25 MeV p xenonl. |
|---|---|---|---|---|
| SCSN-38 | a or a ($\lambda$) | $33 \pm 3$ | | $(18 \pm 1)$ |
| | b or b($\lambda$) | $2.4 \pm 1.7$ | $2.0 \pm 0.2$ | $1.1 \pm 0.1$ |
| Y-7 | a or a($\lambda$) | $5 \pm 1$ | $3.8 \pm 0.5$ | $5 \pm 0.3$ |
| | b or b($\lambda$) | $1 \pm 1$ | $0.4 \pm 0.1$ | $< (0.2 + 0.1)$ |
| K-27 | a or a($\lambda$) | $5 \pm 1.2$ | $4.7 \pm 0.6$ | $5 \pm 0.3$ |
| | b or b($\lambda$) | $0.8 \pm 0.8$ | $0.7 \pm 0.1$ | $< (0.2 + 0.1)$ |

# 3 Series investigations on radiation hardness

The SCI material for the ZEUS calorimeter has been produced in many production cycles (mixtures). To ensure that there are no differences in the radiation stability we have always investigated two samples (dimensions: $200 \times 50 \times 2.6 mm^3$) of 38 production units. They have been exposed in air to 40 Gy from a $^{60}Co$-source with a high dose rate of about 2 kGy/h and measured before and 4 weeks afterwards, when total recovery in air was reached, with the movable xenon lamp. Fig.1 shows the ratio of the light yields after recovery and before irradiation. From this curve the radiation induced additional absorption coefficient $\Delta\mu$ and the loss in light yield can be extracted. We have not tried to understand this curve completely and to develop a formula which describes its (non exponential) behaviour. Our only aim was to compare the different samples. For this we have fitted all the curves in two different regions I and II (where they all could be fitted very well) with the functions:

$$L_i = A_i \cdot (e^{-\Delta\mu_i x} + Re^{-\Delta\mu_i (400mm - x)}) \ , i = \mathrm{I}, \mathrm{II} \tag{2}$$

R = 0.29 is the reflexion factor for an open SCI end. The only two parameters A and $\Delta\mu$ describe the reduced flourescence yield or the additional absorption coefficient.

Fig.2 shows the induced absorption coefficients $\Delta\mu$ for region I (upper curve) and region II. The "error bars" represent the difference of the two samples of one cycle. The variations of the absorption coefficient $\Delta\mu$ between the cycles are comparable to the spreads within the cycles. The mean value and the spread of all cycles are for region I (near the read-out edge):
$\Delta\mu_{\mathrm{I}} = (3.6 \pm 0.4) \cdot 10^{-3} mm^{-1}$
and for region II:
$\Delta\mu_{\mathrm{II}} = (2.3 \pm 0.3) \cdot 10^{-3} mm^{-1}$ for a dose of 40 kGy.
The ratio $b = \Delta\mu_{\mathrm{I}}/D = 0.9 \cdot 10^{-6} cm^{-1} Gy^{-1}$ (see eq.1) is in good agreement with the previous measurements performed with much smaller SCI samples[7].
The radiation-caused reductions in light yield have been calculated by extrapolating the fits of region I (fig.1) to x=0 mm. Here also no essential differences could be seen. The mean loss in fluorescense light yield for all these samples is about $7 \cdot 10^{-4}$% Gy$^{-1}$.

# 4 Dose rate investigations of radiation damage

As pointed out, the high dose rate investigations may underestimate the real radiation damage in the calorimeter[3], where SCI and WLS are exposed to relatively low dose rates . This seems to be especially valid if oxygen is present during the irradiation. Zorn[5] has discussed this item in more detail. We have studied the radiation damage of SCIs and WLSs at medium and high dose rates (30 - 1000 Gy/h) and very low dose rates (2 mGy/h). The results will be presented in sections A and B.

## A Medium and high dose rate exposures

SCSN-38 scintillators (thickness 2.8mm , size$(26 \cdot 2) cm^2$) have been exposed in air to $\gamma$ rays of different $^{60}Co$ sources. The total absorbed dose (D = kGy) was kept constant , while the mean dose rates were 30, 100 and 1000 Gy/h, respectively . The corresponding exposure times were T = 1, 10.5 and 31.5 days.

To detect radiation damage the scintillators were excited using a xenon flash lamp. The fluorescence light was absorbed by a WLS (30 ppm Y7 in PMMA) transmitting the light to a photomultiplier tube. In this case a black coverage at the open end of the scintillator suppressed the reflection of light. The results for irradiations in dry air are presented in fig.4. The light yield ratio $L_{irr}(x)/L_{unirr}$ of irradiated and unirradiated samples has been determined.

Directly after the end of a short term irradiation (fig.3) one observes a strong initial radiation damage. Recovery of polysterene base materials in air is very fast. The permanent radiation damage remaining after (1-2) days storage in air is much weaker than the initial damage. The data plotted in fig.3b,c show that the behaviour during long term irradiations differs considerably from that described above. Samples of 2.8 mm thickness recover during irradiation if the dose rate is less than 100 Gy/h. After the end of the long term irradiations practically no further changes of radiation damage have been observed. A second result is that we find no dependence of the permanent damage on the dose rate within the experimental errors. The important consequence for the scintillator is that the radiation damage expected after long term irradiations in air can be determined from short term irradiations with subsequent recovery in air.

The above statements confirm with higher accuracy the results obtained using a spectrophotometer [4].Quantitatively the observed effects can be described in terms of the diffusion model [11,12]. The radiation creates free radicals which act as absorption centers so that the SCI appears yellow-brown. When the samples are stored in contact with air, the oxygen diffuses into the SCI and forms peroxides which do not absorb visible light. Therefore, for recovery in air after short term high dose rate irradiations, bleached zones with thickness $z$ at both surfaces of the SCI can be observed, where $z$ increases with time after the end of the irradiation due to the subsequent delivery of oxygen from the outside by diffusion. In the case of a thin sample with d = 2.8 mm at dose rates below 100 Gy/h the diffusion delivers enough oxygen to recover nearly all created absorption centers at once, so that the sample is completely transparent (totally recovered) at the end of the irradiation.

## B Low dose rate irradiations

A SCSN-38 SCI of 60 cm length and 5cm width has been irradiated in between DU plates. Together with a shielded (not exposed to DU) sample of the same dimensions, cut out of the same larger plate and machined in one process, it has been fixed via light guides to a photomultiplier. The absorbtion coefficients (AC) and light yields of both SCI have been measured by placing a $^{106}$Ru source always at the two ends of the scintillators. The same set-up was used for pairs of SCI with thicknesses of 2.6 and 5 mm.

As a result fig.4 shows the AC for the 5 mm thick samples over a period of 478 days , corresponding to a totally accumulated dose of 14.3 Gy. Both SCI behave in the same way. The AC increases by about 14% within this period and seem to converge to a constant value of about 1.05 - 1.06 $m^{-1}m$. This increasure apparently is due to normal ageing. Within the errors no radiation effect is to be seen.

Fig.5 shows the AC of the 2.6 mm thick SCI, measured over 1100 days, corresponding to a dose of about 42 Gy. Here the AC of the non-irradiated sample increases only by about 3% while the SCI exposed to DU shows an increase of 9%. If this difference would be caused by the radiation it would be a very large effect. The radiation-induced additional absorption coefficient $\Delta\mu$ would be a factor 20 higher than expected from the high dose rate irradiations (table 1). But there are some hints that this difference is not caused by the radiation: Both curves converge with increasing time to constant nearly identical AC. One might deduce that the non-irradiated sample had a different "pre-ageing" when the measurement started. It is not clear why this should happen but we have seen similiar effects for two identical WLS K-27 in PMMA which were not exposed to any radiation [6]. The fact that both AC of the 5 mm thick samples (fig.6) show a growth (normal ageing) which is similiar to the 2.6 mm irradiated SCI also indicates that the non-irradiated 2.6 mm thick sample behaves differently from the others. Furthermore one would not expect a convergence to a constant AC of the irradiated SCI but a nearly linear increase with time if the effect would be caused by radiation.

The ratios of the light yields of the DU-exposed and non-exposed SCI showed within the errors no changes with time.

For the DU irradiation of WLS K-27 in PMMA we have seen strong changes of the absorption coefficient[6] which probably have to be explained by normal ageing.

No radiation damage effect has been seen when a SCSN-38 plate has been exposed in air to 230 Gy from a $^{90}Sr$-source with a dose rate of 0.25 Gy/h [9]. In contrast to this an "Altulor" SCI already showed a strong damage effect for half the dose [3].

On the other hand studies at the Tevatron collider[10] have shown that there are already strong damages of SCSN-38 and other SCI after an exposure to 150 - 200 Gy with rates of about 0.5 Gy/a.

A possible strong dose rate effect has been seen for the CDF beam counters made of 25.4 mm thick SCSN-23 SCI [8]. SCSN-23 has a twice as high concentration of the first fluor b-PBD and another second fluor (BBOT) compared with SCSN-38.After a one year run of the collider the AL decreased from 109 to 15 cm and the light yield was reduced for 40%. In the first two months of the run the dose unfortunately was not measured, afterwards the SCI have accumulated only 40 Gy. Consequently either there is a strong dose rate effect or the SCI have seen a very strong dose within the first two months of the run.

# 5 Summary

The results can be summarized as follows:

(1) The low term high dose rate (1 - 10 kGy/h) irradiations of SCSN-38 and Y-7 and K-27 up to 24 kGy with subsequent storage in air and inert gases resulted in the same induced permanent absorption coefficients $\Delta\mu$ and $\Delta\mu(\lambda)$ for exposure to $^{60}Co$ or 25 MeV protons. Series investigations on 38 production cycles showed no larger deviations. The reduction in light yield of SCSN-38 was stronger for irradiation in air than in argon. The radiation stability of the investigated materials is high enough for 10 years of ZEUS operation.

(2) At medium dose rates of $\dot{D}$ = 30 - 100 Gy/h in air the SCI SCSN-38 of 3 mm thickness is always in the totally recovered state and the damage can be calculated from the short-term experiments. The damages are higher than expected from the accumulated dose due to the fact

that the diffusion of oxygen into the interior of the material is not fast enough. For the WLS the situation is more complicated and critical due to an initial blocking of the recovery.

(3) At very low dose rates (some mGy/h) in air some experiments show a much stronger damage of SCI and WLS than expected from the applied dose from short-term irradiations. On the other hand it is not totally excluded that these effects are caused by other reasons than radiation, e.g. ageing or chemical reactions of oxygen with shorter lived radicals. Especially the ageing may be responsible for strong changes of the attenuation length.

## 6 References

[1] ZEUS Collaboration,"The ZEUS detector", Status Report 1989, March 1989.

[2] Manufactured by KYOWA GAS CHEMICAL INDUSTRY, see T. Kanon, K. Kondo, A. Yamashita, T. Shimizu, L. Nodulman, "A new scintillator and wavelength shifter", *Nucl. Instr. and Meth.*, vol. 213,(1983) 261

[3] Y.Sirois and R.Wigmans , *Nucl. Instr. and Meth.* vol. 213,(1985) 262.

[4] D.Paul, P.Schröder, V.Stieber, K.Wick, proceedings of the ECFA *Study Week on Instrumentation of High Luminosity Hadron Colliders* ed. E.Fernandez,G.Jarlskog,vol.1(1989) 159

[5] C. Zorn, *IEEE Trans. Nucl. Sc.*, vol. 37 (1990),504

[6] U. Holm, see ref.[4] , p. 163.

[7] U. Holm and K. Wick, *IEEE Trans. Nucl. Sc.*, vol. 36,(1989) 579.

[8] N. Giokaris, private communication

[9] J. Straver and R.Wigmans, private communication

[10] C. S. Lindsey, E. W. Anderson, C. Hojvat, F. Turkot, A. Bujak, *Nucl. Instr. and Meth.*, vol. A 254,(1987) 212.

[11] K.Wick,G.Riedel,D.Paul,V.Stieber, *Nucl. Instr. and Meth.*,vol.A 227,(1989) 251

[12] D.Paul,P.Schröder,V.Stieber,B.Bicken,K.Wick, to be published in *Nucl. Instr. and Meth.*

## 7 Figures

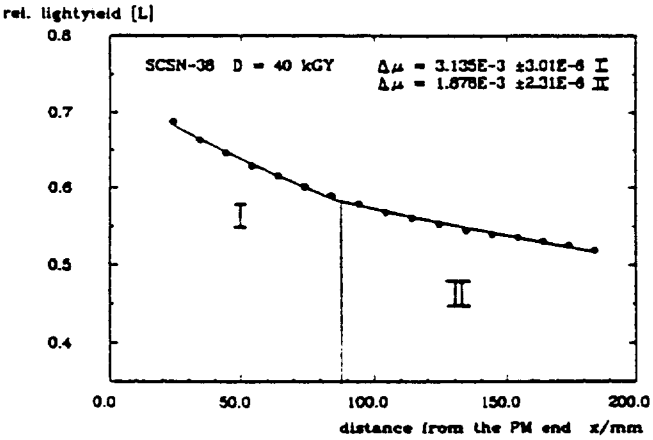

Figure 1: *Light yield ratio, measured with a zenon lamp, of a SCI sample after total recovery and before irradiation. The solid lines represent two fits with the functions of eq.2 in Regions* I *and* II.

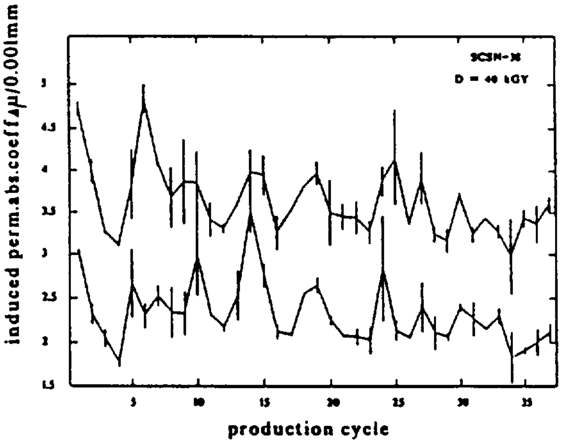

Figure 2: *Radiation induced permanent absorbtion coefficients* $\Delta\mu$ *for region* I *(upper curve) and region* II *for 38 production cycles.*

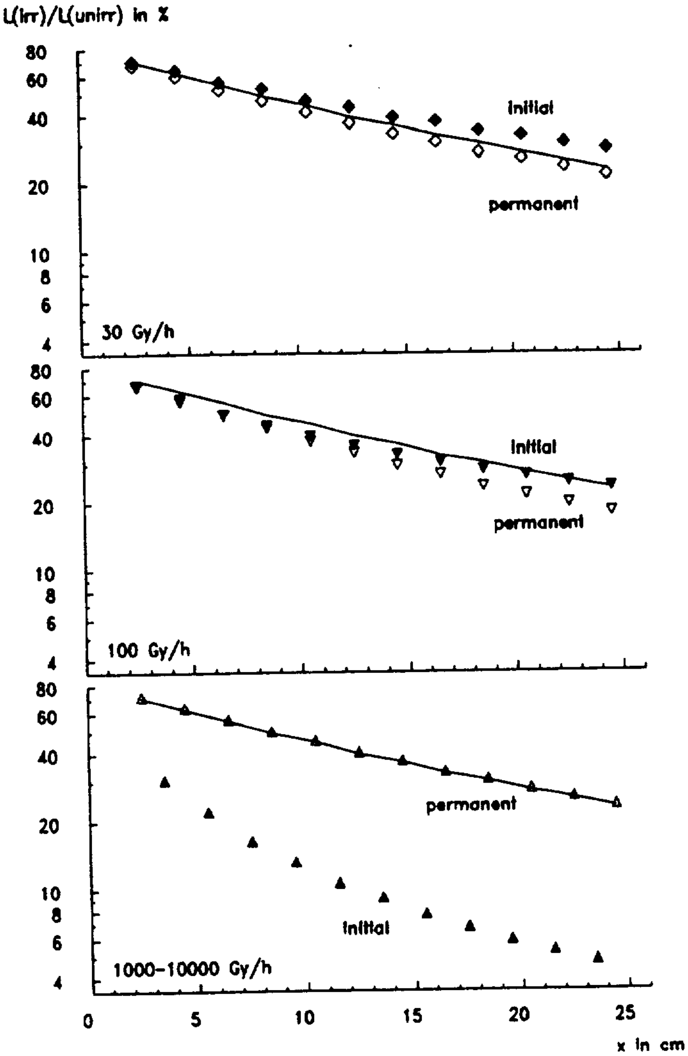

Figure 3: *Light yield ratios of irradiated and unirradiated SCSN-38 scintillators measured after exposure to a fixed dose* $D = 24$ *kGy at different dose rates* $\dot{D}$. *The solid and open symbols refer to measurements performed directly or more than 100 days after irradiation ( initial and permanent radiation damage ).The solid lines indicate the permanent damage measured at a high dose rate.*

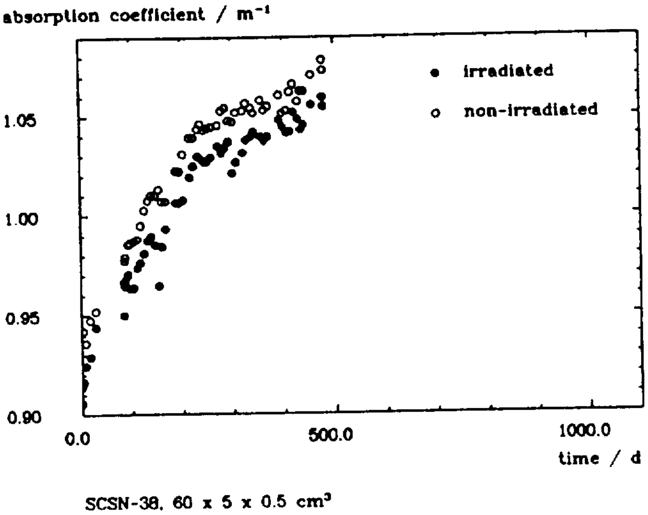

Figure 4: *Absorption coefficients of a SCI SCSN-38 (thickness 5mm), exposed to DU ,as a function of the exposure time.For comparison the AC of an identical non-irradiated sample is shown.*

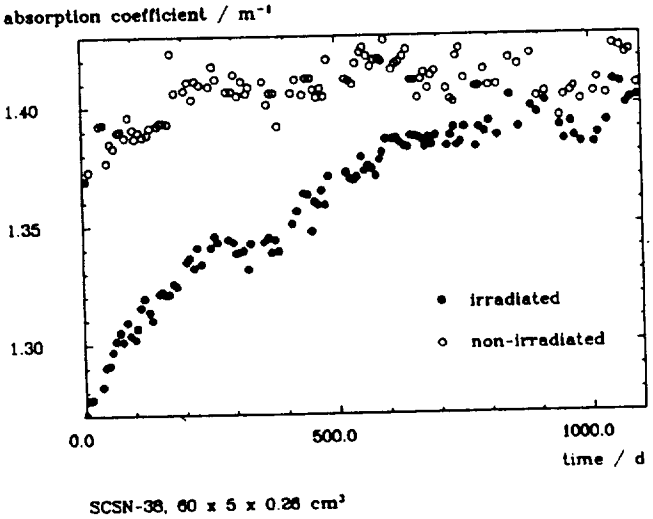

Figure 5: *The same as in fig.4 but the SCI thicknesses are 2.6 mm .*