Neutron/gamma radiation damage comparison of plastic scintillators

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\(^{a}\) Supported by BMBF
1 Introduction

Design of any experimental set-up at high energy physics colliders, especially at very high luminosity proton-proton colliders:

Stability under severe radiation levels?

Radiation damage for plastic scintillators, wave-length shifters, light guides in calorimeters means:

- destruction of fluors $\rightarrow$ reduction of light yield
- additional absorption of fluorescence light $\rightarrow$ reduction of attenuation length

As a consequence the calorimeter shows a reduced performance:

- loss in response $\rightarrow$ change of calibration
- resolution worsening
- worsening of spatial homogeneity

Many investigations on radiation damage have been performed, but mainly $\gamma$ irradiations and only few neutron irradiations.
Problem: "pure" neutron sources with high intensity are rare.
But:
Hadronic and electromagnetic showers produce a strong flux of fast neutrons (LHC: up to $10^{15} \text{n/cm}^2 \cdot \text{year}$) with an energy distribution peaked at about 1 MeV!

It is known that in polymeric materials fast neutrons may cause different "damage" compared to γs with the same released dose.

⇒

We have performed neutron irradiations of plastic materials at:

- GKSS/Geesthacht, 5 MW pool type reactor FRG-1. High γ background, much more thermal than fast neutrons.
- SCK-CEN/Mol, natural uranium reactor BR1. Nearly no γs, thermal n flux ten times higher than fast n flux.
- PTB Braunschweig (german "National Bureau of Standards"), cyclotron, nuclear reaction delivers nearly pure beam of fast neutrons.

This paper deals only with the PTB irradiation.
2 Experiment

a) Irradiations

Irradiated materials:

- Scintillator **SCSN38**, polystyrene base, 2.6 mm thick
- scintillator **SCSN81T2**, -",- , 3 mm
- light guide **GS218**, pure polymethylmetacrylate, 5 mm
- wave-length shifter **Y7**, PMMA base, 2 mm

SCSN, Y7 → Kuraray, GS218 → Röhm.
All samples 10 x 10 mm².
Irradiations and recovery in air.

Neutron irradiation:

- Physikalisch Technische Bundesanstalt PTB, Braunschweig; \(^9\text{Be} \ (d,n) \ ^{10}\text{B}\) reaction with 13.2 MeV deuterons from cyclotron → fast neutrons.

- Neutron field (facility for neutron therapy dosimetry) is calibrated to high accuracy (2.5%) by time-of-flight methods and ionization chambers.

- KERMA averaged mean neutron energy for tissue: \(\overline{E}_n = 6\) MeV

- Low \(\gamma\) background (< 5%).
The PTB Facility for Neutron Therapy Dosimetry


Physikalisch-Technische Bundesanstalt (PTB), D-3300 Braunschweig

Introduction

At the PTB, a facility (1) for fast neutron absorbed dose determination is available which is well suited for calibration and investigation of neutron dosimeters, and for international intercomparisons of neutron absorbed dose standards (2). The facility comprises an energy-variable compact cyclotron for high proton and deuteron currents which are used to produce intense collimated neutron beams via the p + Be and d + Be reactions.

Table 1

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Experimental parameters</td>
<td></td>
</tr>
<tr>
<td>Reference position (free in air), 80 cm from the</td>
<td></td>
</tr>
<tr>
<td>target, 0 degrees</td>
<td></td>
</tr>
<tr>
<td>Deuteron energy on the Be target</td>
<td>13.35 MeV</td>
</tr>
<tr>
<td>Deuteron current on the Be target</td>
<td>25 µA</td>
</tr>
<tr>
<td>Field size in the ref. position</td>
<td>10 cm * 10 cm</td>
</tr>
<tr>
<td>Total dose rate in A-150 plastic</td>
<td>5 mGy/s</td>
</tr>
<tr>
<td>Fluence-averaged mean neutron energy</td>
<td>5.24 MeV</td>
</tr>
<tr>
<td>Kerma-averaged mean neutron energy</td>
<td>6.02 MeV</td>
</tr>
</tbody>
</table>

Properties of the neutron field

The spectral neutron fluence, \( \Phi_E \), per unit deuteron beam charge, \( Q \), on the Be target (thickness 2 mm) has been determined by time-of-flight (TOF) techniques using the pulsed deuteron beam of the cyclotron and the fast timing and n-\( \gamma \) discrimination properties of an NE213 scintillation detector (3,4,5). The influence of the target assembly (water cooling and stainless steel backing, each of 1 mm thickness) and of the neutron collimator has been calculated using the Monte Carlo code COLLII (6) (see Fig. 1).

Typical experimental conditions are shown in Table 1.
Samples have been irradiated for 9 hours at three positions (distances to Be target) with **n fluences**

- (I) $1.55 \times 10^{14} n/cm^2$
- (II) $1.45 \times 10^{14} n/cm^2$
- (III) $1.35 \times 10^{14} n/cm^2$

**\( \gamma \) irradiation:**

There are many "old" data on \( \gamma \) induced damage.

But for direct comparison additional irradiation with \( ^{60}Co \) source at Hahn-Meitner-Institut Berlin:

- samples cut from the same plate
- \( \gamma \) dose: 7.0 kGy
- \( \gamma \) dose rate about the same as for n irradiation

**b) Dosimetry**

In addition to the good knowledge of the n field simultaneous irradiation of TLD700 **thermoluminescence** dosimeters and **alanine dosimeters**.

**TLD700 \( \gamma \) doses:**

- (I) 347 Gy
- (II) 364 Gy
- (III) 382 Gy
Alanine dosimeters "Elcugray" (Kabelmetal Electro):

- Cables of 4.8 mm diameter composed of DL-α-alanine and ethylenepropylene rubber.
- Radiation causes stable radicals measured by electron spin resonance methods.
- Alanine dosimeters are calibrated in pure γ-fields. Response to fast neutrons is approximately only half of the γ sensitivity for the same doses.

(Katsumura et al., Radiat. Phys. Chem. 28(1986)33)

Fast neutrons cause nuclear reactions in plastic materials which mainly are composed of H, C, N and O atoms. The main contribution of the neutron energy release in material is **elastic scattering** of neutrons, especially by hydrogen atoms:
recoils have low energy, high dE/dx → **high LET** (linear energy transfer).

Doses corresponding to n fluence calculated via **KERMA** factors (kinetic energy released in material).

KERMA factors for elements (at 6 MeV; Attix, Intr. to Radiol. Physics ...):

\[
\begin{align*}
\text{H: } & \quad 4.17 \cdot 10^{-10} \text{Gy/n/cm}^2 \\
\text{C: } & \quad 7.82 \cdot 10^{-12} \text{Gy/n/cm}^2 \\
\text{N: } & \quad 1.15 \cdot 10^{-11} \text{Gy/n/cm}^2 \\
\text{O: } & \quad 4.96 \cdot 10^{-12} \text{Gy/n/cm}^2
\end{align*}
\]
KERMA factors for irradiated materials:

Alanine \( (C_3H_7NO_2) \): \( 3.96 \cdot 10^{-11} \text{ Gy/n/cm}^2 \)
PMMA \( (C_5H_8O_2) \): \( 3.97 \cdot 10^{-11} \text{ Gy/n/cm}^2 \)
PS \( (C_8H_8) \): \( 3.96 \cdot 10^{-11} \text{ Gy/n/cm}^2 \)

At 6 MeV these values are accidentally nearly identical!

The neutron doses at the different irradiation positions are for all four materials:

(I): 6.15 kGy,  (II): 5.76 kGy,  (III): 5.36 kGy

Together with the \( \gamma \) doses one gets the total doses:

(I): 6.53 kGy,  (II): 6.12 kGy,  (III): 5.71 kGy

Alanine, irradiated at position (III):

n dose 5.36 kGy, 50\% n efficiency \( \Rightarrow \) expected signal
2.68 kGy + 0.35 kGy = 3.03 kGy
measured: 2.96 kGy \( \Rightarrow \) very good accordance, 50\% response for fast neutrons confirmed.
c) Spectrophotometry

Transmittance measurement with spectrophotometer:

\[ T(\lambda) = T_0(\lambda) \cdot e^{-\mu(\lambda) \cdot d} \]

Radiation damage:

\[ \Delta \mu(\lambda) = \mu_{irr}(\lambda) - \mu_{non}(\lambda) \]

Radiation induced changes of transmission partly very small at doses 6-7 kGy

⇒ high precision measurements needed (e.g. reproducibility of wave-length setting 0.1 nm).
3 Radiation damage and recovery

Irradiation of polymers (here polystyrene (PS) and polymethylmethacrylate (PMMA)) →

formation of radicals $R\cdot$

which strongly absorb (fluorescence) light. Radicals often are rather stable (at room temperature) but they react violently with oxygen ($R\cdot + O_2$) →

formation of peroxy radicals $RO_2\cdot$

which do no more absorb. Transmission has recovered to a remaining generally much smaller permanent damage.

Optical damage of PMMA even may increase after end of irradiation before recovery:

"build up" effect

Little oxygen $\Rightarrow R\cdot + R\cdot \rightarrow A,$
where $A$ shows an even stronger absorption than $R\cdot$. 
\[ \Delta \mu(D, t) = a \cdot D \cdot f(t) + b \cdot D \]

with:  
\[ a = c - b, \quad c : \text{initial damage/dose}, \]
\[ b : \text{permanent damage/dose} \]

\[ f(t) = \begin{cases} 
1 & \text{for } t = 0 \\
0 & \text{for } t > t_E 
\end{cases} \]

- initial and permanent damages increase linear with dose D (at least up to 100 kGy)
• recovery depends on amount of oxygen:

  – PS samples (SCSN) irradiated to 10 kGy recover in about two days
  – PMMA samples (GS218, Y7) need more than a year if air of normal pressure and room temperature is around.

**PS** samples during measurement always in totally recovered state.

**PMMA** samples have been measured both in initial and in permanent damage state. Total recovery by "baking" for one hour at 115 °C.
SCSN38--Transmission

**Neutrons**

$D_{\text{total}} = 6.5 \text{ kGy}$

**Gammas**

$D_{\gamma} = 7.0 \text{ kGy}$
SCSN38--neutron induced absorption coefficient $\Delta \mu (n)$

$\Delta \mu_n \left(420\text{nm}\right) = 0.052 \text{ cm}^{-1}$
SCSN38--gamma induced absorption coefficient $\Delta \mu (\gamma)$

$\Delta \mu_{\gamma} (420 \text{nm}) = 0.011 \text{ cm}^{-1}$

"Older" values 0.013
Results

SCSN38--ratio $\Delta \mu (n) / \Delta \mu (\gamma)$

$\lambda / \text{nm}$

emission peak SCSN38
SCSN38--neutron induced absorption coefficient $\Delta \mu (n)$

$D_{\text{total}} = 6.5$ keV

SCSN38--gamma induced absorption coefficient $\Delta \mu (\gamma)$

$D_{\gamma} = 7.0$ keV

Destruction of BDB fluors, stronger for $\mu$s
SCSN81T2--neutron induced absorption coefficient $\Delta \mu (n)$

$D_{total} = 6.5 \text{ kGy}$

$\Delta \mu_n (425 - \text{um}) = 0.038 \text{ cm}^{-1}$
SCSN81T2--gamma induced absorption coefficient $\Delta \mu (\gamma)$

$\Delta \mu (435 \text{ nm}) = 0.012 \text{ cm}^{-1}$
SCSN81T2--gamma induced absorption coefficient $\Delta \mu (\gamma)$

$D_{\gamma} = 7.016 \gamma$

$\Delta \mu_{\gamma} (435 \text{ nm}) = 0.012 \text{ cm}^{-1}$
SCSN81T2--ratio $\Delta \mu (n) / \Delta \mu (\gamma)$

influence of dye destruction

emission peak SCSN81T2
GS218--Transmission

\[ D_{total} = 6.1 \text{ bGy} \]

\[ D_{total} = 7.0 \text{ bGy} \]

non-irradiated
non-irradiated, baked
irradiated, recovered by baking
irradiated, 14d after irr.

initial Transm.

\( \lambda / \text{nm} \)

\( \nu \text{s} \)

\( \text{gammas} \)

\( \lambda / \text{nm} \)
GS218--Transmission

**neutrons**

$D_{total} = 6.4 \text{ Gy}$

- non-irradiated
- non-irradiated, baked
- irradiated, recovered by baking
- irradiated, 14d after irr.

**initial Trauma.**

**gammas**

$D_{total} = 7.0 \text{ Gy}$

- non-irradiated
- non-irradiated, baked
- irradiated, recovered by baking
- irradiated, 19d after irr.

**initial Trauma.**
GS218--initial absorption coefficients $\Delta \mu (\gamma)$ and $\Delta \mu (n)$

- Gamma irradiation $D = 7.0 \text{ kGy}$
- Neutron irradiation $D = 6.1 \text{ kGy}$
GS218--initial absorption coefficients $\Delta\mu (\gamma)$ and $\Delta\mu (n)$

$D_\gamma = 7.0 \text{ kGy}$
$D_{\text{total}} = 6.1 \text{ kGy}$
Y7--initial absorption coefficients $\Delta \mu (\gamma)$ and $\Delta \mu (n)$
Number of destroyed Y7 dyes:
\[ \Delta \mu^* (460 \, \text{nm}) = -0.003 \, \text{mm}^{-1} \]
\[ \mu (\cdot) = 0.432 \, \text{mm}^{-1} \]
Y7--initial absorption coefficients $\Delta \mu (\gamma)$ and $\Delta \mu (n)$

$D_\gamma = 7.0 \text{ kGy}$

$D_{\text{total}} = 6.5 \text{ kGy}$

- $\gamma$-irradiation
- $n$-irradiation

$\lambda / \text{nm}$

Y7 emission

Y7 emission peak
Fig. 1. The spectra of scintillator output light.

<table>
<thead>
<tr>
<th>Scintillator</th>
<th>Doped materials</th>
<th>Stabilizer</th>
</tr>
</thead>
<tbody>
<tr>
<td>SCSN38</td>
<td>1% b-PBD 0.02% BDB</td>
<td></td>
</tr>
<tr>
<td>SCSN38T2</td>
<td>1% b-PBD 0.02% BDB</td>
<td>0.02% stabilizer</td>
</tr>
<tr>
<td>SCSN61T2</td>
<td>1% PHB 0.02% BDB</td>
<td>0.02% stabilizer</td>
</tr>
<tr>
<td>SCSN71</td>
<td>1% PHB 0.02% BSB</td>
<td></td>
</tr>
<tr>
<td>SCSN71T2</td>
<td>1% PHB 0.02% BSB</td>
<td>0.02% stabilizer</td>
</tr>
<tr>
<td>SCSN71T5</td>
<td>1% PHB 0.02% BSB</td>
<td>0.05% stabilizer</td>
</tr>
<tr>
<td>SCSN71T5B a</td>
<td>1% PHB 0.02% BSB</td>
<td>0.05% stabilizer</td>
</tr>
<tr>
<td>SCSN81T2</td>
<td>1% PHB 0.02% TBBT</td>
<td>0.02% stabilizer</td>
</tr>
<tr>
<td>SCSN81T2B a</td>
<td>1% PHB 0.02% TBBT</td>
<td>0.02% stabilizer</td>
</tr>
</tbody>
</table>

a These scintillators are different from others for their temperature.
4 Summary

- Scintillators SCSN38, SCSN81T2 (polystyrene based), pure polymethylmetacrylate GS218 and wavelength shifter Y7 in polymethylmetacrylate have been irradiated in a fast neutron beam. ($E_n \approx 6$ MeV)

- n field very well known, only little $\gamma$ background. In addition alanine and TLD dosimetry.
  n fluences: $1.5 \cdot 10^{14} n/cm^2$ corresponding to neutron doses of 6 kGy (via KERMA factors).

- For comparison $\gamma$ irradiation of samples from same plate.
  $\gamma$ dose: 7 kGy.

- Accurate measurement of radiation induced optical absorption coefficient:
  Polystyrene based SCSN38 (SCSN81T2) is a factor 5 (3) more sensitive to fast neutrons than to $\gamma$s. The number of destroyed fluor molecules is smaller for neutrons.

- PMMA materials GS218 and Y7 are less sensitive to fast neutrons than to $\gamma$s. The same holds for the destruction of Y7 molecules.

- Irradiations with higher n fluences are in progress.