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EFFECTS OF NUCLEAR RADIATION ON THE
OPTICAL PROPERTIES OF CERIUM-DOPED GLASS

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ABSTRACT

Some twenty types of glass containing 0.5-4% CeO_2 have been irradiated in a ^{60}Co gamma cell and in the mixed neutron-gamma field of a nuclear reactor, at total integrated doses of up to 5×10^9 rad (CH). The resulting colouration has been assessed quantitatively by measuring the light transmission with reference to air, in the range 360-510 nm. From the results, certain types of glass suitable for applications in nuclear engineering can be selected. Specifically, it was found that 1-2% CeO_2 content is usually sufficient to obtain radiation-resistant optical glass: the reduction in light transmission above 450 nm is nil at 10^8 rad (CH), below 10% at 10^9 rad (CH), and below 20% at 5×10^9 rad (CH); the post-irradiation fading is negligible.

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1. INTRODUCTION

Glass is used in accelerators for various applications, for example viewing windows in vacuum chambers, TV camera objectives, insulation of RF cavities, electron tubes, etc., or for dosimetry.

Colour changes are produced in normal glass during exposure to ionizing radiation. This colouration, which starts around 5×10^5 rad (CH) *) and is very dark at 10^7 rad (CH), can either

- a) hamper operation if it occurs in the glass components of systems where maximum optical transmission is essential, or
- b) be utilized for radiation dose measurements (dosimetry).

Therefore, in order to avoid case (a) above, types of radiation-resistant glass have been developed which overcome the colouring effects produced by radiation. Glass can be protected from colouring by the addition of certain polyvalent ions called "protective agents" of which cerium is the best known. Addition of 1 to 2% CeO_2 suppresses the radiation-induced colour in cerium glass due to the Ce^{4+} ion, which is such a powerful electron acceptor that it removes the radiation-created free electrons in the matter which would otherwise form colour centres.

In this report we present the radiation test results of some types of cerium-doped glass studied, which could be of interest for nuclear engineering. The glass samples have been irradiated in a ^{60}Co gamma cell and in a nuclear reactor up to a total integrated dose of 5×10^9 rad (CH).

2. MATERIALS, TEST SAMPLES AND TEST METHODS

The types of glass which have been studied were manufactured by Jenaer Glaswerke Schott, Germany ($45 \times 10 \times 2.5$ mm³) and by Chance-Pilkington, England ($30 \times 10 \times 10$ mm³ and $30 \times 10 \times 5$ mm³). Table 1 shows the types of glass and their CeO_2 content, which varies between 0.5 and 4%.

Each sample of glass was prepared with the 45×10 mm² or 30×10 mm² faces highly polished, flat and parallel.

In order to assess the colouration induced by radiation, the percentage change in spectral transmission in the near ultraviolet/visible region of the electromagnetic spectrum was measured using a Beckman DB-GT spectrophotometer. Measurements were made against air as reference. The glass samples were scanned over the spectrum from about 510 to 360 nm with a scan speed of 5 nm/min. Measurements from 510 to 800 nm showed no difference in spectral transmission and have therefore been omitted. The direct linear transmission recordings were made on a Beckman potentiometric recorder.

*) 1 rad (CH) = energy absorption of 100 erg/g in $(\text{CH})_n$ -material (e.g. polyethylene).

Table 1

Types of glass and reduction of light transmission at 1×10^8 , 1×10^9 , and 5×10^9 rad (CH) ($\lambda = 450$ nm).
(Manufacturer: Schott, except Nos. 9, 10 Pilkington.)

No.	Glass type	CeO ₂ Content	Reduction of light transmission ($\lambda > 450$ nm) at dose of			Figure
			1×10^8 rad (CH) *)	1×10^9 rad (CH) **)	5×10^9 rad (CH) **)	
9	RSF	0.5	84	86	95	1, 11
16	SF 19 G7	0.7	13	26	30	2
18	SF 8 G7	0.7	11	12	12	
19	SF 1 G7	0.7	0	22	21	
3	WG 9 G9	0.9	4	20	29	3, 12
12	SK 10 G10	1.0	10	23	37	
5	BAK 1 G12	1.2	2	12	13	4
14	F 2 G12	1.2	3	5	6	
15	SF 16 G12	1.2	3	3	12	
8	SK 4 G13	1.3	7	28	49	5, 1
1	BK 7 G14	1.4	2	13	21	
7	LF 5 G15	1.5	0	1	6	
17	LAK N9 G15	1.5	18	64	74	
10	OW 10	1.7	14	40	54	6
11	F 2 G20	2.0	0	1	3	
2	BK 7 G25	2.5	0	13	16	7
4	GG 375 G34	3.4	1	8	24	8
6	LF 4 G34	3.4	3	11	15	
13	F 6 G40	4.0	2	9	11	9

*) Reactor irradiation position Ebene 1

**) Reactor irradiation position 11

Table 2

Characteristics of radiation sources and total doses to glass samples

Irradiation source	⁶⁰ Co γ cell	Nuclear reactor		
		SNIF	Position 11	Ebene 1
Fast neutron flux ($E > 1$ MeV) [n_f/cm^2 sec]	-	$3 \times 10^9 - 5 \times 10^9$	$2 \times 10^{12} - 3 \times 10^{12}$	$2 \times 10^{10} - 3 \times 10^{10}$
Thermal neutron flux [n_{th}/cm^2 sec]	-	$1 \times 10^9 - 2 \times 10^9$	$4 \times 10^{12} - 5 \times 10^{12}$	$3 \times 10^{11} - 4 \times 10^{11}$
γ dose rate [rad (CH)/h]	1×10^6	$\sim 1 \times 10^4$	$1 \times 10^8 - 2 \times 10^8$	1×10^7
Irradiation temperature [°C]	20	~ 30	~ 30	~ 30
Irradiation medium	air	air	water	air
γ dose to samples [rad (CH)]	5×10^6 1×10^7 5×10^7 1×10^8		1×10^9 5×10^9	1×10^8 5×10^8 1×10^9
Fast neutron fluence to samples [n_f/cm^2]*)	-	2.5×10^{14} 7×10^{14}		

*) $10^{16} n_f/cm^2 = 1 \times 10^9$ rad (CH)

3. IRRADIATION CONDITIONS

The irradiations were carried out in a ^{60}Co gamma cell and in three different irradiation positions, i.e. SNIF, Ebene 1, and Position 11 of the ASTRA Reactor near Vienna. Table 2 summarizes the radiation sources, their characteristics, and the doses with which the glass samples have been irradiated. Further details about the irradiation facilities, dosimetry methods, etc., can be found in Schönbacher et al.¹⁾.

4. RESULTS

Since many of the specially developed types of glass listed in Table 1 show similar results, we only present in this report those types which show characteristic behaviour, for example those with low and high alteration of optical transmission after irradiation. A complete summary of all the test results is given in a report by McGrath²⁾.

The results are presented in the form of graphs, which are self-explanatory, namely:

- i) Figures 1 to 9: change of optical transmission as a function of wavelength.
- ii) Figure 10: change of optical transmission as a function of CeO_2 content.
- iii) Figures 11 and 12: post-irradiation fading effects.

In Table 1 we summarize the reduction of light transmission at 1×10^8 rad (CH), 1×10^9 rad (CH), and 5×10^9 rad (CH) ($\lambda = 450$ nm).

5. ANALYSIS OF RESULTS AND CONCLUSIONS

- i) Among the tested samples, types of glass have been found which
 - are not at all affected by nuclear radiation up to 1×10^8 rad (CH),
 - show a reduction in light transmission of less than 10% at 1×10^9 rad (CH) ($\lambda > 450$ nm),
 - show a reduction of light transmission of less than 20% at 5×10^9 rad (CH) ($\lambda > 450$ nm).
- ii) A CeO_2 content of 1.0-2.0% seems, in general, to be sufficient to obtain radiation-resistant optical glass. A CeO_2 content higher than 2% does not necessarily improve the properties of a glass under irradiation (see Fig. 10).
- iii) The colouration of the glass depends on the irradiation conditions. The damage parameter is greater with neutrons than with ^{60}Co γ -irradiation for glass exposed to identical dose levels.
- iv) The post-irradiation fading effect of the glass at room temperature two months after irradiation is negligible, whereas heating the glass for several hours at 300°C anneals the created colour centres (see Figs. 11 and 12).
- v) The light transmission with reference to air of samples of unirradiated cerium-doped glass is 80 to 90% (e.g. they appear slightly yellowy brown).

REFERENCES

- 1) H. Schönbacher, M. Van de Voorde, A. Burtscher and J. Casta, Study of radiation damage to high-energy accelerator components by irradiation in a nuclear reactor, Kerntechnik 17, 268 (1975).
- 2) B. McGrath, Compilation of results of light transmission measurements on various types of cerium-doped glass, Internal report LAB II-RA/TM/75-34 (1975).

Figs. 1 to 9 Change of optical transmission as a function of wavelength for different radiation doses and sources. Reference medium: air. Radiation doses [in rad (CH)]: (1) unirradiated glass; (2) 2.5×10^5 ; (3) 7×10^5 ; (4) 5×10^6 ; (5) 10^7 ; (6) 5.5×10^7 ; (7) 1.1×10^8 ; (8) 10^8 ; (9) 5×10^8 ; (10) 10^9 ; (11) 10^9 ; (12) 5×10^9 . The irradiations were done as follows: (2) and (3), at SNIF; (4) to (7), with Co-60; (8) to (10), at Ebene 1; (11) and (12), at Position 11.

Fig. 1 9-Pilkington type RSF
CeO₂ content: 0.5%

Fig. 2 16-Schott type SF 19 G7
CeO₂ content: 0.7%

Fig. 3 3-Schott type WG 9 G9
CeO₂ content: 0.9%

Fig. 4 5-Schott type BAK 1 G12
CeO₂ content: 1.2%

Fig. 5 8-Schott type SK 4 G13
CeO₂ content: 1.3%

Fig. 6 10-Pilkington type OW 10
CeO₂ content: 1.7%

Fig. 7 2-Schott type BK 7 G25
CeO₂ content: 2.5%

Fig. 8 4-Schott type GC 375 G34
CeO₂ content: 3.4%

Fig. 9 13-Schott type F 6 G40
CeO₂ content: 4%

Fig. 10 Change of optical transmission as a function of CO_2 content. Reference medium: unirradiated glass. Wavelength: 410 nm.

Fig. 11 Post-irradiation fading effects in 9-Pilkington type RSF glass. Reference medium: air. (1) Unirradiated; (2) 5.5×10^8 rad (CH); (3) 4 h in light at room temperature; (4) $2\frac{1}{2}$ h at 100°C ; (5) $2\frac{1}{2}$ h at 100°C + 1 h at 200°C ; (6) $2\frac{1}{2}$ h at 100°C + 4 h at 200°C ; (7) $2\frac{1}{2}$ h at 100°C + 4 h at 200°C + 4 h at 300°C .

Fig. 12 Post-irradiation fading effects in 3-Schott type WG 9 G9 glass. Reference medium: air. (1) Unirradiated; (2) 5.5×10^8 rad (CH); (3) $2\frac{1}{2}$ h at 100°C + 4 h at 200°C ; (4) $2\frac{1}{2}$ h at 100°C + 4 h at 200°C + 4 h at 300°C .