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Methodology for Prediction of Near-Earth Space Radiation Effects on Optical Glasses

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Prediction of near-Earth space radiation impact on spectral transmission of optical glasses remains an issue after several decades of research. The very wide range of multicomponent commercial glasses, radiation types, total doses, doses rates, etc. question the possibility of ab initio assessment of radiation impact. It has been proposed that a pragmatic parametric approach has merit in such a situation. We use BK7 glass as an example to demonstrate how the problem can be approached. Based on a sequence of transmission spectra measured in the course of and after gamma-irradiation (Co^{60} source), we show how to find the required parameterization set. Taking into account the number of optical glasses used in space optical instruments, it is proposed that the first step toward a useful radiation effects database consists of the elaboration of a uniformly applicable methodology for testing of optical glasses. We believe that our work can serve as a basis for that methodology.

Nomenclature

A_i	=	amplitude of ith absorption band, cm^{-1}
a_{0i}	=	maximal (saturated) absorption, cm^{-1}
c	=	dose coefficient for defect generation, rad^{-1}
D	=	total dose, rad
D_s	=	saturating dose, rad
i	=	index indicating ith defect type
k_a	=	rate of defect annealing, s^{-1}
k_g	=	rate of defect generation, s^{-1}
N	=	number of absorption bands
n	=	total instantaneous concentration of defect precursors, m^{-3}
n_i	=	instantaneous concentration of i-th type defect precursors
n_0	=	total initial concentration of defect precursors, m^{-3}
n_{0i}	=	initial concentration of ith type defect precursors, m^{-3}
$q(t)$	=	relaxation function
R	=	dose rate, rad/s
s_i	=	proportionality coefficient between the absorption amplitude and the defect concentration, cm^{-1}/m^{-3}
t	=	time, s
α	=	index of the fractional exponent
α_{Di}	=	dose coefficient, $cm^{-1}rad^{-1}$
Δa	=	induced absorption coefficient, cm^{-1}
Δa_i	=	induced absorption coefficient for ith band, cm^{-1}
σ_i	=	bandwidth of ith absorption band, eV
τ_0	=	characteristic relaxation time, s
$\varphi(k)$	=	relaxation rates distribution, s
ω_{0i}	=	peak position of ith absorption band, eV

Introduction

RADIATION impact on space optical instruments operating in the near-Earth radiation environment remains a concern. Many investigations have been undertaken over the past few decades, and a significant insight into the underlying physical effects has been gained.[1,2] Nevertheless, for the designer and user of space optical instrumentation, the question of qualification of optical materials for use in a space radiation environment remains an issue. A common practice is that radiation hardness of a new system is addressed by conducting new tests. The results obtained for particular materials and systems lie scattered throughout the literature and are difficult to use because of the very specific nature of the tests carried out. The very wide range of materials, radiation types, total doses, dose rates, and parameters actually measured makes any extrapolation doubtful and the application of the data to a new situation rarely possible. In addition, the experimental tests as presented in the literature are not always completely documented and not easily accessible or understandable to a nonspecialist.

Radiation influences various characteristics of optical glasses; the most important are transmission, refractivity, and density.[3–5] Variation of these characteristics during and after irradiation has been found to be very sensitive to both material and irradiation conditions. Taking into account the number of possible irradiation scenarios and the amount of experimental work to be done in each case, it would be very attractive to simulate such experiments based on first principle concepts. Unfortunately, such an approach has never been demonstrated. Moreover, the complicated chemical composition of commercial optical glasses and the complexity of radiation–matter interaction question that approach despite the significant progress in computation technologies. Recent work in this field has shown that there is merit in a pragmatic parametric approach.[6] In the present work, we analyze a parameterization scheme intended for description of radiation-induced transmission degradation. It is proposed to use this scheme for establishing a database for assessment

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of the radiation impact on spectral transmission of optical glasses.

Analysis of Radiation-Induced Transmission Degradation

Transmission degradation under ionizing radiation is a consequence of the generation of charge carriers (electrons and holes). Localization of those charges in traps leads to creation of radiation defects. Such defects correspond to electronic states, which are not present in the glass before irradiation. Optical transitions, which are related with those states, give rise to an additional absorption in the whole spectral range from the infrared to the UV. A decrease in the density of initial electronic states corresponds to a decrease in absorption in the region of initial absorption bands. However, such induced transparency is usually not of interest because, before exposure to radiation, glasses are transparent in the working spectral range.

In general, an electronic state in glassy material can be converted to another one by radiation. The exact nature of radiation defects depends on the chemical composition of glass and is highly sensitive to the presence of trace impurities. The most susceptible to radiation are dangling bonds, oxygen bridges, overcoordinated atoms, that is, deviations from the "ideal" random network. The concentration of such radiation precursors is usually much lower than the concentration of normal bonds, and the saturation effect plays an important role.

Radiation-induced defects in glass can be thermally, optically, or radiation transformed to other configurations. The concentration of radiation-induced defects changes after the end of irradiation; some of them can be completely annealed, but most defects are metastable and are characterized by very long relaxation times.

In the present model we consider defect generation only via the first-order reactions. The evolution kinetics of the precursor's concentration is described by the following differential equation, which takes into account generation, annealing, and the saturation effect:

$$\frac{\mathrm{d}n}{\mathrm{d}t} = -k_g n + k_a(n_0 - n) \tag{1}$$

The first term describes the decrease of concentration caused by transformation of precursors to metastable defects, and the second one takes into account the defect annealing.

We assume also that the rate of defect generation is proportional to the radiation dose rate R:

$$k_g = cR \tag{2}$$

where the constant c depends on material characteristics only. This assumption remains realistic until the defect concentration remains small in comparison with the concentration of normal states, a situation relevant for space applications. The constant c can be analytically computed based on known models of defect generation. However such computations are rather complicated, and the accuracy is not very high. A more realistic approach is to find c using experimental data.

It follows from Eq. (1) that the defect concentration n_i shows a saturating behavior:

$$n_i(k_a) = n_0[k_g/(k_a + k_g)][1 - \exp(-[k_a + k_g]t)] \tag{3}$$

Here we have introduced the index i in order to take into account that different types of hole and electron color centers are present in irradiated glass. Those defects are characterized with different spectroscopic parameters, generation, and relaxation rates.

The solution shows that at the initial stage the defect concentration grows linearly, and it saturates with increase of irradiation time. The saturation level depends on both the generation and the annealing rates. If relaxation is very fast ($k_g \ll k_a$), the saturation level is significantly smaller than the level defined by the precursor concentration. In the absence of relaxation ($k_a = 0$), we can rewrite Eq. (3) as

$$n_i = n_{0i}[1 - \exp(-cD)] \tag{4}$$

which makes it evident that c is the inverse of the saturating radiation dose D_s.

In disordered materials relaxation is characterized by a broad distribution of the relaxation rates $\varphi(k)$ with parameters different for each defect type:

$$n_i = \int \mathrm{d}k_a \varphi_i(k_a) n_i(k_a) \tag{5}$$

The amorphous nature of glass results also in an inhomogeneous broadening of defect-related optical transitions, which are well characterized as Gaussian bands.[7] The induced absorption coefficient Δa_i is proportional to the defect concentration. Therefore

$$\Delta a_i(\omega) = A_i \exp\left[\frac{-(\omega - \omega_{0i})^2}{2\sigma_i^2}\right], \qquad A_i = s_i n_i \tag{6}$$

The observed absorption spectrum is a superposition of absorption bands:

$$\Delta a(\omega) = \sum_{i=1}^{N} A_i \exp\left[\frac{-(\omega - \omega_{0i})^2}{2\sigma_i^2}\right] \tag{7}$$

We have now obtained a set of formulas, which allow the induced absorption under radiation or after the end of irradiation to be described. The practical application of this approach requires the kinetic parameters defining defect generation (n_0, c) relaxation rates distribution [$\varphi_i(k_a)$], along with spectroscopic parameters (ω_{0i}, σ_i) to be known for each type of defect.

From the practical point of view, it is more convenient not to use (n_{0i}, c_i) but the parameter pair

$$a_{0i} = s_i n_{0i}, \qquad \alpha_{\mathrm{D}i} = c_i n_{0i} \tag{8}$$

where a_{0i} is the maximal (saturated) absorption in the absence of relaxation and $\alpha_{\mathrm{D}i}$ is the dose coefficient, which describes the initial (linear) part of the defect accumulation. In the next section we show how these parameters can be found from the optical transmission measurements.

Experimental Results and Discussion

We have selected BK7 (Schott) glass to illustrate the presented approach. The samples were polished disks 0.2 mm thick. The optical transmission spectra were recorded in the spectral range from 200 to 800 nm using a commercial double-beam spectrophotometer. The natural logarithm of the ratio of the transmission before irradiation to that after irradiation, normalized by the sample thickness, gives the radiation-induced absorption coefficient.

A Co^{60} source with a dose-rate of 6 krad/h (water) at the sample's location was used for γ irradiation of the samples (1 rad corresponds to 10^{-2} J of energy absorbed by 1 kg of material, the SI unit of absorbed dose is the gray = 100 rads). Irradiation was performed in a stepwise dose-accumulation manner. First the samples were irradiated with a dose of 400 krad. Optical measurements were performed with a 2-h delay. Then the samples were irradiated again, so that the total accumulated dose reached 800 krad. After the end of irradiation, transmission spectra were measured several times in a time interval up to 1700 days in order to monitor postradiation relaxation. In total, we have 12 meaningful spectra. Several transmission spectra measured on a 0.2-mm thick BK7 glass sample are shown in Fig. 1.

To construct the kinetic curves, it is necessary to resolve the absorption spectra into individual absorption bands. As a result of nonorthogonality of the Gaussians, such decompositions are not unique, and additional information is required to obtain a physically meaningful result. Based on the data available in the literature, we assume that for relevant radiation loads the experimental spectra in the range 230–800 nm can be described with four bands.

The selection of the number of the bands is a very important question, and it cannot be solved automatically. Moreover, a good fit does not guarantee that the selected bands are physically present.

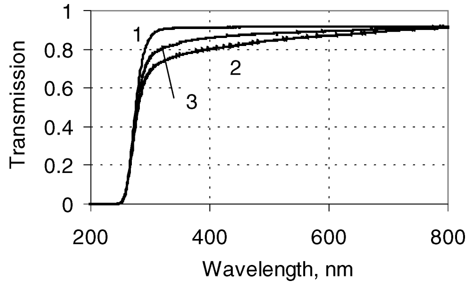

Fig. 1 Transmission spectra measured on a 0.2-mm-thick BK7 glass sample: 1, before irradiation; 2, after 800-krad dose; and 3, 1619 days of annealing at standard laboratory conditions.

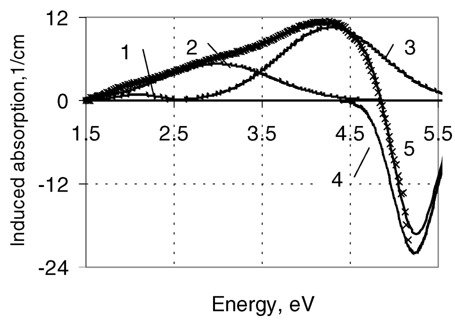

Fig. 2 Decomposition of the induced absorption spectra for BK7 glass sample into four Gaussian bands: 1, 2.09-eV band; 2, 3.0-eV band; 3, 4.28-eV band; 4, 5.23-eV band; 5, sum; □, experimental data; and D = 800 krad.

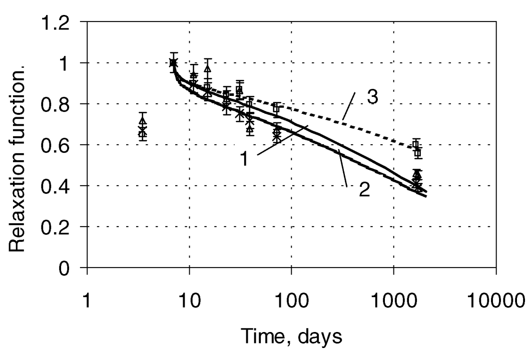

Fig. 3 Variation of the amplitudes of the absorption bands: □, △, ∗, measured data; ——, fit with the stretched-exponential relaxation functions; line 1 and △, 2.09-eV band; line 2 and ∗, 3.00-eV band; and line 3 and □, 4.28-eV band.

An indirect confirmation that the decomposition is correct is that the physical functions give a good description of the relaxation kinetics.

We have used a two-step procedure to find the absorption band parameters from the experimental data. First, we fit each spectrum independently. We assumed that the central energy and the width of those bands are changed neither during the experiment nor after the end of irradiation. This assumption is supported by the results of the fit, which show a small scattering of the values supposed to be constant. In the next step only the amplitudes of the absorption bands were allowed to vary. All other values were taken as the average of the values obtained in an independent fit of each induced absorption spectrum. An example of the decomposition is shown in Fig. 2.

In this way we have computed the variation of the absorption bands amplitudes in the course of and after irradiation, as shown in Fig. 3.

Relaxation in strongly correlated systems is usually nonexponential. Indeed, we have found that the Debye-type (exponential) relaxation does not describe our experimental results. In contrast,

Table 1 Set of parameters required for the description of the radiation impact on BK7 optical glass samples

Band no.	ω_{0i}, eV	σ_i, eV	$\alpha_{\mathrm{D}i}$, cm krad^{-1}	a_{0i}, cm^{-1}	τ_0, days	α
1	2.09	0.245	3.53E−03	1.51	3,061	0.322
2	3.00	0.628	1.99E−02	11.3	2,024	0.291
3	4.28	0.587	4.10E−02	26.4	16,200	0.260
4	5.23	0.246	−8.40E−02	−75.0	2,043	0.229

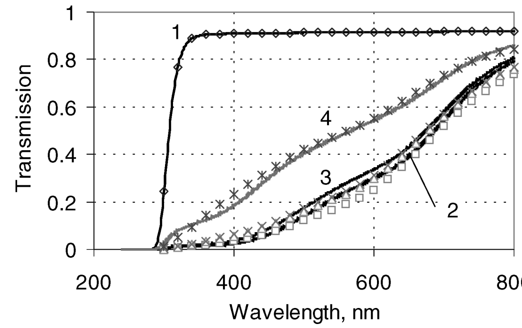

Fig. 4 Comparison of the simulated (△, □, ∗, ◇) and measured (curves) results obtained on a 5-mm-thick BK7 glass sample. Irradiation during seven days up to the dose of 800 krad: 1, before irradiation; 2, just after irradiation; 3, four days after irradiation; and 4, 1619 days after irradiation.

the stretched-exponent (Kohlrausch) law

$$q(t) = \exp[-(t/\tau_0)^\alpha], \qquad 0 < \alpha < 1 \tag{9}$$

gave a reasonably good description for the whole interval up to 1700 days of annealing. Simple analytical expressions for the relaxation rates distribution, corresponding to Eq. (8), are known only for a few values of α. Therefore, the distribution was computed by numerical inversion of the Laplace transform, which in not always convenient. The problem of the relaxation function selection is still to be addressed.

The results are summarized in Table 1. It must be explicitly stated that Table 1 is given here as an example and is directly applicable only for dose rates similar to those in our experiment.

The data in Table 1 form a set of parameters that provide a quantitative description of transmission change of BK7 glass under radiation. Figure 4 compares simulation and experimental results for a 5-mm-thick sample, which was irradiated up to 800 krad during seven days. A good agreement is observed. Only three first bands were used in the simulation because the fourth band (5.232 eV) gives no contribution for wavelengths longer than 300 nm. In addition, the values measured for that UV band are not reliable because of the very strong intrinsic absorption of BK7 in the UV below 280 nm. The approximation error is somewhat higher at 800 nm, which indicates that extension of the measurement range for longer wavelengths is also required.

Database Approach

Essential steps in the design of an optical system intended for use in a space radiation environment are schematically outlined in Fig. 5. The goal of this procedure is to estimate the end-of-life performance. If it does not satisfy the requirements, the optomechanical configuration/design is changed, and the procedure is repeated.

The absence of easily available data in a standard format is a problem when necessary lifetime predictions must be done urgently and at a minimal cost. A recent example has been qualification assessment of the optical system of the Fluid Science Laboratory (FSL) being constructed for flight on the European laboratory module (Columbus) of the International Space Station. The total absorbed dose expected at the FSL optical bench level is predicted to be 700 rad, but a qualification level of 1400 rad was specified for safety. Common engineering sense and experience leads one to

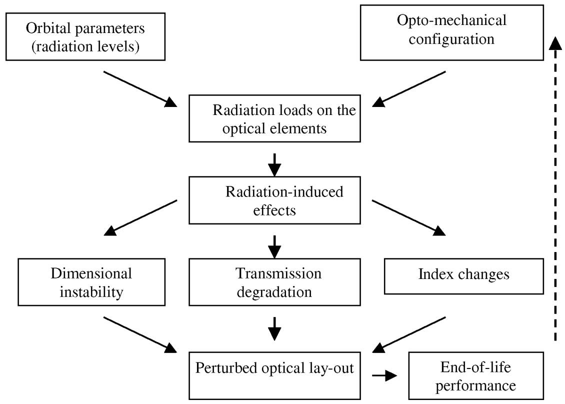

Fig. 5 Schematics of a radiation-tolerant optical system design process.

expect an insignificant radiation-rmance degradation at such low dose levels accumulated over a 10-year on-orbit lifetime. Nevertheless, it was decided to proceed with an expensive testing of the full set of glasses because otherwise reliable data, which would allow quantifying the effect of radiation, are not available. It is obvious that the radiation testing of the FSL-relevant glasses has to be performed under conditions specific for its qualification. However, the absence of a standard and a generic approach makes it rather probable that the same glasses will have to be tested again for another mission with different radiation conditions.

The optical glass industry is passing now through a transition period, when traditional, well-established types of glasses are replaced with new types.[8] It can be expected that new glasses will completely replace, in the near future, the presently used glasses. At the same time, the previously available so-called "radiation-tolerant" (cerium doped) glasses are being discontinued from routine production. The new glasses are designed to have optical properties in the visible identical to those of the glasses they replace but with higher chemical stability and lower density. This is achieved via changes in the chemical composition, which can result in a significantly different response to radiation. The radiation hardness of these new glasses has not been addressed up to now. This problem must also be dealt with as use of these glasses in space optical systems becomes more widespread.

Conclusions

We have developed a new methodology to parameterize optical glass transmission measurement data for its eventual use for radiation tolerance estimation. The first results obtained with this methodology look very promising. However, a more detailed analysis of several important issues is still required. One of those questions is the influence of the radiation type, that is, will it be sufficient to introduce a scaling coefficient to describe the effect of proton (electron) radiation based on the gamma-irradiation data or will a new set of parameters be required for each radiation type?

Establishing a comprehensive and reliable database for assessment of the radiation impact on spectral transmission of optical glasses requires a very significant amount of experimental work to be undertaken. Taking into account the number of optical glasses used in space optics and possible changes in the nomenclature, it is proposed that a realistic approach calls for the introduction of a widely accepted and uniformly applicable methodology, such as proposed here, for testing of optical glasses. It would be particularly advantageous to introduce this methodology as an industrial standard (ISO and European Cooperation for Space Standardization, perhaps), which can be made applicable for radiation qualification testing of all optical glasses performed under ESA contracts. Test data would be deliverable to the Agency in a suitable format. In this way the Agency will be able to accumulate experimental data required to build, maintain, and update the database on a continuous basis.

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