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Neutron and gamma irradiated optical property changes
for the final optics of the National Ignition Facility
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ABSTRACT

Based on studies we have performed with several radiation sources such as pulsed nuclear reactors, we have been able to construct a physical picture and measure quantitative parameters necessary to model the radiation-induced losses expected for fused silica and fused quartz National Ignition Facility (NIF) target area. It is important to note that these surrogate radiation sources do not have identical temporal and spectral characteristics to NIF, therefore caution is in order since the results obtained to date must be extrapolated somewhat to predict NIF performance.

1. INTRODUCTION

We are currently pursuing an experimental project to understand and quantify the effect that neutron and gamma radiation from the NIF targets will have on the transmission of the final optics assembly (fused silica, KDP/KD*P, and mirror coatings). We are focusing on the penetrating radiation (gamma-rays and neutrons) that will propagate through the debris shield and supply a dose of ~2 krad from neutrons and ~1 krad from gamma rays for each 20 MJ equivalent NIF shot.¹ Much progress has been made by developing a physical model of the damage mechanisms and quantifying the relevant parameters on surrogate radiation sources such as a pulsed nuclear reactor and a continuous ⁶⁰Co gamma ray source.²⁻³ The remainder of this paper will focus on fused silica for which we have the most data and the most complete understanding.

2. EXPERIMENTAL DATA AND ANALYSIS

Our results have shown that both gamma-rays and neutrons are expected to have a measurable impact on the transmission properties of SiO₂. Although 25 types of SiO₂ were studied, these samples led to only four characteristic radiation-induced absorption spectra as shown in Fig. 1. From this information and data obtained on other radiation sources, the peak at 246 nm was determined to arise from direct neutron collision-induced displacements creating a localized oxygen deficient center (ODC) that is intrinsic to SiO₂; the absorption peak at 300 nm (B1 band) is ascribed to arise from the conversion of the ODC defect absorption that is present prior to irradiation from the manufacturer. The peak at 210 nm (E' defect) is due to a two step process whereby the 246 nm ODC defect is first created and then γ-rays convert the ODC into a E' center. The peak centered at 550 nm is most likely due to Al impurities in the SiO₂ as suggested in the literature. The four spectra shown in Fig. 1 correspond to four classes denoted FS1, FS2, FQ1, and FQ2, where FS = fused silica and FQ = fused quartz.

Using the information in Fig. 1 along with data obtained on other radiation sources, we have been able to put together a physical model of the primary radiation damage pathways for fused silica and fused quartz as illustrated in Fig. 2.

Figure 1. Under identical 200 krad neutron and 460 krad gamma irradiation doses, the absorption spectra of all 25 types of SiO₂ studied fell into four distinct classes as illustrated in parts (a), (b), (c), and (d). Parts (a) and (c) correspond to radiation resistant and non radiation resistant fused silica designated FS1 and FS2. Parts (b) and (d) correspond to radiation resistant and non-radiation resistant fused quartz designated FQ1 and FQ2.

Figure 2. Mechanistic model of primary radiation damage mechanisms in SiO₂ that lead to changes in optical performance. Note that both neutrons and gamma rays either separately or in combination can lead to radiation induced damage.

In an effort to benchmark the effective lifetime of the NIF optics we found that it was necessary to utilize a mixed neutron and gamma-ray source, since there are synergistic neutron and gamma-induced pathways that lead to radiation damage. The SNL pulsed reactor number 3 (SPR-III) has provided us with our most relevant radiation source to date, with 100 μ s pulse operation, a mixed neutron and gamma dose ratio that is similar to NIF (within a factor of 5), and a gamma spectrum that is very close to that of NIF. The neutron spectrum from this largely unmoderated U_{235} fission reactor, however, is broad and extends from keV to ~ 3 MeV energies with a peak flux at 1 MeV. This is low as compared to the direct DT 14 MeV neutrons that will dominate the NIF spectrum. As a consequence these results must be extrapolated to higher neutron energies where more damage is likely per individual collision, for making NIF predictions. Figure 3 shows the results of performing a dose dependent study on SPR-III over the course of two days. In one year with 385 MJ of fusion yield, NIF is predicted to produce ~ 40 krads of neutron dose and ~ 20 krads, hereafter referred to as a "NIF year". Note that broken down by radiation type, one shot on SPR-III corresponds to one NIF year for neutrons and 5 NIF years for γ -ray.

Figure 3 presents the results of exposing the best fused silica and fused quartz (part a and b respectively) with up to eight shots on the SPR-III reactor. Note that with fused silica (part a), the absorption peak at 246 nm grows most rapidly at first and at higher doses the 210 nm absorption peak growth rate overtakes it. This qualitative observation is consistent with the model described by Fig. 2.

Figure 3. Optical absorption spectra of fused silica (a) and fused quartz (b) taken after successive shots on the SNL pulsed nuclear reactor number III. One shot has 40 krads neutrons and 100 krads γ -ray dose.

Quantitative modeling for the defect absorption peaks shown in Fig. 3a for fused silica can be performed by applying rate equations derived from the model in Fig. 2a for the number density of neutron-induced ODCs and subsequent γ -ray induced E' defects denoted as N_{ODC_n} and $N_{E'}$ respectively. The rate equations can now be written as:

$$\frac{dN_{ODCn}}{dt} = M_{col}\sigma_{col}(E)N_{SiO_2}\frac{dF_n}{dt} - \beta_E\frac{dD_\gamma}{dt}N_{ODCn}, \text{ and} \quad (1)$$

$$\frac{dN_E}{dt} = \beta_E\frac{dD_\gamma}{dt}N_{ODCn}. \quad (2)$$

The first term in Eq. 1 describes the formation of ODC defects due to direct neutron collisions where F_n is the neutron fluence, σ_{col} is the neutron collision cross section, N_{SiO_2} is the silica number density, and M_{col} is a collision multiplier that denotes the number of cascade collision-induced defects following a single primary neutron impact as show in the first step of Fig. 2a. Equation 2 and the corresponding last term in Eq. 1 accounts for the second step in Fig. 3a where ODC defects are transformed into E' defects with γ -rays based on a phenomenological conversion coefficient denoted β_E , and D_γ is the γ -ray dose in krads. Defect optical absorption cross sections (σ_{ODC} and $\sigma_{E'}$) are then multiplied by the resulting number densities obtained from Eqs. 1 and 2.

For the modeling of the defect absorption peaks shown in Fig. 4b for fused quartz the direct conversion of as-manufactured ODC defect into B1 defects, with number densities N_{ODCm} and N_{B1} respectively, as described by Fig. 2b can be written as:

$$\frac{dN_{B1}}{dt} = -\frac{dN_{ODCm}}{dt} = \beta_{B1}\frac{dD_\gamma}{dt}N_{ODCm} \quad (3)$$

where β_{B1} is the gamma ray conversion coefficient, and there is a non-zero initial condition for the as-manufactured ODC defect concentration determined from absorption spectra of the individual samples.

The open points in Fig. 4 gives the peak defect absorption coefficients obtained from Lorentzian curve fits to the spectra in Fig. 3. The solid lines are fits to the model described above by Eqs. 1-3 with parameters obtained from these curve fits given in Table I.

Figure 18
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Figure 4. Radiation-induced absorption of E' (210 nm), ODC (246 nm), and B1 (300 nm) bands in SiO₂ for fused silica type FS1 and fused quartz type FQ1 in parts (a) and (b), respectively. Data denoted as open symbols was obtained from SNL pulsed nuclear reactor experiments. The numerical modeling using Eqs. 1-3 is denoted as solid lines.

Table I. Parameters utilized to model data from SPR-III reactor and to predict NIF final optics performance.

Parameter	Value	Units	Source
Neutron absorption coefficient, $N_{\text{SiO}_2}\sigma_{\text{col}}$	0.35	cm^{-1}	literature
ODC to E' γ -ray conversion coefficient, $\beta_{\text{E'}}$	0.0007	s/krads	data fit
Collision multiplier for 1 MeV neutrons, M_n	105	unitless	data fit
Collision multiplier for 14 MeV neutrons, M_n	200	unitless	other data
ODC to B1 γ -ray conversion coefficient, β_{B1}	0.004	s/krad	data fit
Optical cross section for ODC defect, σ_{ODC}	$2.1\text{e-}18$	cm^2	data fit
Optical cross section for E' defect, $\sigma_{\text{E'}}$	$3.2\text{e-}17$	cm^2	literature
Optical cross section for B1 defect, σ_{B1}	$1.8\text{e-}18$	cm^2	data fit

To first order, the two unknown optical cross sections, σ_i , and the and the neutron multiplier, M_n , determine the height of the four curves, while the two conversion coefficients, β_i , determine the shape of the data curves.

In order to apply this model to NIF-like conditions, the neutron multiplier obtained at 1 MeV average neutron energy must be extrapolated to 14 MeV neutron energies for the DT fusion neutrons that will be present on NIF. We have analyzed one piece of data on fused silica taken in 1987 with 14 MeV DT neutrons on the LLNL rotating target neutron source number II (RTNS-II). The spectral shape is virtually identical to that shown in Fig. 3a for the fission reactor induced absorption, indicating that no fundamentally new defects are created. The number of defects per collision is expected to be higher for 14 versus 1 MeV neutrons, however. By fitting the RTNS spectra with the model described above, we have inferred that the collision multiplier is ~ 200 for 14 MeV neutrons. The error bar is large (factor of 2 to 5) on this number since the details of the radiation doses (specifically ratio of neutron to gamma dose) are uncertain. Further experiments are planned on other high energy neutron radiation sources to validate this preliminary assessment with well-controlled irradiation conditions. Nevertheless, with the parameters determined from the experiment in Table I, Eqs. 1-3 can be utilized to predict the NIF performance of the final SiO_2 optical components.

Figure 5 presents the results of this modeling with both fused silica type FS1 and fused quartz type FQ1, with an initial condition of $N_{\text{ODC}_m} = 3 \times 10^{16} \text{ cm}^{-3}$ determined from data. A DT fusion yield of 385 MJ/year is presently assumed for the NIF and will be the baseline case for the computations here. The final focus lens is assumed to be 4.5 cm thick. Thicker lens designs would be expected to have larger absorbance (-log transmission) which is proportional to the thickness. It is apparent from the predictions in Fig. 5 that FS1 fused silica is significantly more radiation resistant than FQ1 fused quartz at 3ω . Fused silica is expected to have $>99\%$ transmission over the 30 year facility lifetime. Fused quartz, however is expected to drop to a 99% lens transmission within one year. If the shot rate or target yields on NIF were increased by 2 to 4

Figure 5. Predicted transmission of NIF final focus lens versus time for fused silica type FS1 (solid lines) and fused quartz type FQ1 (dashed line). Predicted transmissions for the assumed NIF fusion yield of 385 MJ/year as well as 2X and 4X that shot rate as shown for fused silica.

times (yield rates of 770 and 1540 MJ/yr.), the lifetime of the silica final focus lenses would be reduced. Figure 5 presents our predictions for these shot rates which lower the effective lifetime to 18 and 9 years for 2X and 4X increase in yield rate, respectively.

3. Conclusions

For the baseline fused silica expected to be used on NIF, the final lens transmission is expected to exceed 99% over the 30 year NIF facility lifetime, although if fused quartz were chosen the transmission is expected to drop to 99% within one year. If the shot rate or target yields on NIF were increased from the assumptions of 385 MJ/yr. to 770 and 1540 MJ/yr., the 99% transmission lifetime of the silica final focus lenses would be reduced somewhat to 18 and 9 years, respectively, which should be workable.

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