

Optical bleaching in situ for barium fluoride crystals

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Received 12 September 1994

Abstract

This report presents results of optical bleaching for barium fluoride crystals to be used to construct a precision electromagnetic calorimeter at future hadron colliders. A practical technique for in situ bleaching by illuminating crystals with light from a UV lamp through an optical fiber is established. The light attenuation length of current production BaF_2 crystals can be set to around 200 (160) cm under 400 (130) rad/h by using 4.3 (1.1) mW bleaching light from a mercury (xenon) lamp through a \emptyset 0.6 mm fiber. The color center dynamics model proposed in our early publication [1] has also been verified. The technical approach described in this report can be used for other candidate crystals to maintain an adequate light attenuation length in a radiation environment.

1. Introduction

In last few years, extensive research and development has been carried out to investigate the feasibility of building a crystal electromagnetic calorimeter in future hadron colliders [2]. The clear physics potential of this precision electromagnetic calorimeter in searches for the Higgs boson in intermediate mass region attracted physics community pursuing the origin of the electroweak symmetry breaking [4-7]. At the time of this writing, a crystal detector is actively being considered as an option for the CMS electromagnetic calorimeter at the Large Hadron Collider (LHC), and research and development for various crystals is very active in this area [3]. A fundamental issue for such a detector, however, is the performance of crystals in a radiation environment with a dose rate up to few hundreds rad/h [6]. Studies showed that the radiation damage and the consequent light response nonuniformity would destroy the energy resolution of a crystal calorimeter [8]. A radiation-hard crystal, or an approach to cure the radiation damage in situ, such as the optical bleaching, thus is crucial for such a detector.

Our previous study [1] on optical bleaching of barium fluoride (BaF₂) crystals shows that a blue light of 400 nm is effective in removing the radiation damage and to reset production size (25 cm long) crystals to a light attenuation length (LAL) of longer than 180 cm. Based on color center dynamics model proposed [1], it was estimated that the required light intensity to set the LAL of BaF₂ crystals to a

The result presented in this report is a continuation of our previous study. In this investigation, we established a practical approach to implement optical bleaching in situ. It was found that a light attenuation length of 160 (200) cm can be achieved for production size BaF_2 crystals under 130 (400) rad/h by using bleaching light through a $\emptyset 0.6$ mm fiber from a 150 (200) W xenon (mercury) lamp. In addition, the color center dynamics model proposed in our previous work [1] was also verified. These results thus established a practical approach to use BaF_2 crystals of current production quality in future hadron colliders, or in any severe radiation environment.

Section 2 summarizes background information: performance of BaF_2 under radiation and the model of color center dynamics proposed in our early work [1]. Crystal samples and the experimental apparatus used in this investigation are described in Section 3. Our results are presented in Section 4.

2. BaF₂ crystals

2.1. BaF₂ performance under radiation

The BaF_2 is a mature crystal – high quality, large size (up to 35 cm long) crystals can be mass produced by manufacturers: the Shanghai Institute of Ceramics (SIC) and the Beijing Glass Research Institute (BGRI). Since its surface can be well polished, the light attenuation length of

stable value of 150 cm (in a dynamic equilibrium) is of an order of mW/cm², or a maximum of 150 W for the entire BaF₂ calorimeter, designed for the GEM collaboration [5], at a luminosity of 10^{33} cm⁻² s⁻¹.

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a BaF_2 crystal can be calculated by measuring its transmittance [9]. The performance of a BaF_2 crystal in a radiation environment has been thoroughly investigated in the last few years, as documented in Ref. [8]. A brief summary is given below.

- There is no permanent damage in BaF_2 caused by dose from photons, neutrons or hadrons. At room temperature, the recovery of the damage is extremely slow. However, all damage recovers fully after a thermal annealing at 500°C for 3 h in a dry atmosphere without oxygen. UV light is also found to be effective in removing damage.

- The radiation damage of BaF_2 shows clear saturation. Both transmittance and light output are stabilized after initial dosage of a few tens to 100 krad. The damage has no dose rate dependence.

– The damage of BaF_2 is caused by the formation of color centers and consequent self-absorption of the scintillation light. There is no damage to the scintillation mechanism.

– The damage mechanism is understood. Impurities (rare earths), defects (inclusions), oxygen and OH^- (U and O^- substitutional centers) are responsible.

2.2. Color center dynamics

The improvement of the intrinsic radiation resistance of BaF_2 , however, is a very difficult process. The light attenuation length of production size (25 cm) BaF_2 crystals after 1 Mrad dosage is found to be 41 cm, i.e. less than the 95 cm specification [8]. Following BaF_2 Expert Panel's recommendations [10], an alternative approach of using optical bleaching in situ was studied [1]. By using a calibrated light source from a monochromator, a blue light of 400 nm was found to be effective in removing color centers, while a light of 500 nm or longer is not.

A color center dynamic model [1] was proposed to explain the optical bleaching. If both annihilation (bleaching) and creation (damage) processes exist, for one kind of color center, we have

$$dD = -aID dt + (D_{all} - D)bR dt, \qquad (1)$$

where D is the optically bleachable color center density in units of m⁻¹, a is the annihilation constant in units of $cm^2mW^{-1}h^{-1}(mW^{-1}h^{-1} \text{ or } J^{-1}$ for fiber bleaching), I is the light intensity in mW cm⁻² (mW or J h⁻¹ for fiber bleaching), D_{all} is the total density of traps related to the optically bleachable color centers in the crystal, b is the damage constant in units of krad ⁻¹, R is the radiation dose rate in units of krad h⁻¹, and t is the time in h. The solution of Eq. (1) is

$$D = D_0 e^{-(aI+bR)t} + \frac{bRD_{all}}{aI+bR} [1 - e^{-(aI+bR)t}], \qquad (2)$$

where D_0 is the initial value of the bleachable color center density. For each value of I and R, an equilibrium between annihilation and creation will be established for one

Table	1
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400	nm	light	intensity	needed t	0	maintain	BaF ₂	at	LAL	==	150	cm
		<u> </u>					_					

η	0	1	2.5	
R [krad/h]	0.02	0.04	0.4	
$[mW/cm^2]$	0.21	0.42	4.2	

kind of color center at an optical bleachable color center density (D_w) of

$$D_{\rm w} = \frac{bRD_{\rm all}}{aI + bR} \,. \tag{3}$$

According to this dynamic model, the intensity of a 400 nm light, required to restore the LAL to a stable value of 150 cm (in a dynamic equilibrium), can be estimated to be 10.5R mW/cm² for production of BaF₂ crystals. Table 1 lists the expected radiation dose rate at the front surface of a BaF₂ calorimeter and the corresponding bleaching light intensities required to set LAL = 150 cm at different rapidities, assuming the BaF₂ calorimeter has a barrel of 75 cm radius and two end caps at $z = \pm 150$ cm. A maximum of 150 W is needed to bleach the entire BaF₂ calorimeter at the standard SSC luminosity (10³³ cm⁻² s⁻¹).

3. Experiment

3.1. Samples

Three tapered 25 cm BaF_2 crystals produced by SIC were used in this investigation. Table 2 lists the dimension, transmittance (*T*) at 220 nm and corresponding light attenuation length (LAL) for these three crystals, where the subscripts 0 and 1M refer to before and after 1 Mrad ⁶⁰Co γ -ray irradiation. The progress in radiation hardness of BaF_2 crystals produced by SIC is clearly shown in the increase of T_{1M} and LAL_{1M}. In some measurement, an additional crystal SIC401, which is 20.5 cm long and has a similar quality as the SIC402, was also used.

All surfaces of the crystal samples were wrapped with two layers of aluminum foil to maintain a stable surface reflection under irradiation, except the large end of the crystal which was coupled to a Hamamatsu R4406 triode. In some measurements, four layers of PTFE teflon (38 μ m thick) film plus one layer of aluminum foil (for protection) were used.

Table 2 Properties of three 25 cm BaF₂ crystals

ension [cm]	<i>T</i> ₀ [%]	LAL ₀ [cm]	Т _{1М} [%]	LAL _{1M} [cm]	
25×3^2	86.1	557	44.5	36	
25×4^2	87.2	779	43.3	34	
$\times 25 \times 4.6^2$	86.1	557	48.6	41	
	ension [cm] 25×3^2 25×4^2 $\times 25 \times 4.6^2$	$\begin{array}{c c} \text{ension [cm]} & T_0 \\ [\%] \\ \hline 25 \times 3^2 & 86.1 \\ 25 \times 4^2 & 87.2 \\ \times 25 \times 4.6^2 & 86.1 \\ \end{array}$	$\begin{array}{c c} \text{ension} [\text{cm}] & T_0 & \text{LAL}_0 \\ [\%] & [\text{cm}] \\ \hline \\ 25 \times 3^2 & 86.1 & 557 \\ 25 \times 4^2 & 87.2 & 779 \\ \times 25 \times 4.6^2 & 86.1 & 557 \\ \hline \end{array}$	$\begin{array}{c c c c c c c c c c c c c c c c c c c $	$\begin{array}{c c c c c c c c c c c c c c c c c c c $

3.2. Transmittance measurement

The transmittance of 25 cm BaF_2 crystals was measured by using a double beam, double monochromator UV/Visible spectrophotometer (Hitachi U-3210) with a large sample compartment equipped with an integrating sphere which is coated with custom Halon. The systematic uncertainty in repeated measurements of transmittance is around 0.2%.

The measured transmittance was converted to the light attenuation length according to Ref. [9]. The bleachable color center density was then defined as the difference between 1/LAL and $1/LAL_0$, where LAL_0 , corresponding to unbleachable color centers, is the light attenuation length after a full bleaching. Note, the LAL_0 is a function of the spectrum of the bleaching light.

3.3. In situ bleaching setup

All in situ bleaching measurements were carried out at JPL's radiation facility which is a radiation chamber equipped with a 14000 Ci⁶⁰Co source. Fig. 1 shows the setup used for in situ bleaching experiment. A crystal and a dosimeter were placed in a light sealing black box. The dose rate was adjusted by placing the black box in different location in the radiation chamber, and by changing the shielding used. The whole volume of entire crystal was uniformly irradiated.

The crystal's large end was coupled to a Hamamatsu R4406 triode through a thin layer of optical grease (Dow Corning 200). The R4406 has a solar blind photocathode (K-Cs-Te) which suppresses the response to the light with wavelength longer than 300 nm [11]. The anode dc current of R4406 was monitored to give an indication of



Fig. 2. Schematic view of bleaching light propagation in a BaF_2 crystal in the test setup used at Caltech, where light directly from fiber (primary light) has an open angle of 60°, while the reflected light (secondary light) covers the whole volume of the crystal.

crystal's transmittance during irradiation. 2 m monitoring R4406 anode dc current and high voltage, and a monitor for photofeedback unit were placed outside the radiation chamber.

The bleaching light was produced by a UV lamp: either a 150 W xenon lamp (Oriel 6254) or a 200 W mercury lamp (Oriel 6283) was used. The power supply of the UV lamp was controlled by a photofeedback unit (Oriel 68850) which provides a light intensity stability of around 0.2%. The light was coupled to a 2 m \emptyset 0.6 mm optical fiber (Oriel 77517). The fiber was coupled to the test crystal at a corner of the large end face which was coupled to R4406. In some measurements, a dichroic reflector (Oriel 66219) with a bandpass of 420 to 680 nm was placed between the fiber and the UV lamp to reduce the light intensity in useless long wavelengths (> 500 nm). Fig. 2 is a schematic showing the light propagation in the crystal. Because of the 60° opening angle of the light cone and the reflection of the crystal surface, the entire crystal was uniformly illuminated by the fiber light.



Fig. 1. Setup for in situ bleaching at JPL's radiation chamber, using a 150 W xenon (or a 200 W mercury) lamp and a \emptyset 0.6 mm fiber. The entire volume of the BaF₂ crystal was uniformly irradiated.



Fig. 3. Schematic view of a bleaching setup used at Caltech, where light was shot through a monochromator to the back face of the BaF_2 crystal.

In some measurements, bleaching light from a monochromator (Bausch & Lomb, Grating 1200 Grove/mm, 10 nm bandwidth) was used. Fig. 3 shows a schematic of the setup. The light with a rectangular spot of 0.8 in. (height) $\times 1$ in. (width) was shot to the large end of the sample. No reflector was placed at either end of the crystals. Typical intensity was determined to be mW/cm², depending on the wavelength used [1].

3.4. Calibration of the light intensity

The intensity of the bleaching light was calibrated by measuring the photocurrents of a UV enhanced silicon detector (Oriel 70116). First, the total photocurrent, $I_{\rm si}^{\rm tot}$, was measured by shooting the light through the fiber directly to the Si detector. Second, the light through the fiber was shot through a 1/8 M monochromator (Oriel 77250), which is driven by a stepping motor (Oriel 77325), to a 6 in. integrating sphere (Oriel 70451). The photocurrent as a function of wavelength, $I_{\rm si}(\lambda)$, was measured by the Si detector which was also coupled to the integrating sphere. The differential light intensity, $I(\lambda)$, from the fiber as a function of the wavelength, λ , can be deduced as

$$I(\lambda) = \frac{I_{\rm si}^{\rm tot}I_{\rm si}(\lambda)}{E_{\rm g}(\lambda)RS_{\rm si}(\lambda)\int \frac{I_{\rm si}(\lambda)}{E_{\rm g}(\lambda)}d\lambda},$$
(4)

where $E_{g}(\lambda)$ and $RS_{Si}(\lambda)$ are the efficiency of the monochromator system and the radiant sensitivity of the detector. Both are provided by the vendor (Oriel). We further define an integrated light power, $P(\lambda_{cut})$, as a function of the cut-off wavelength, λ_{cut}

$$P(\lambda_{\rm cut}) = \int_0^{\lambda_{\rm cut}} I(\lambda) \, \mathrm{d}\lambda.$$
⁽⁵⁾

Since our early tests showed that only the light with a wavelength shorter than 500 nm is effective in bleaching, the bleaching light power is defined as the integrated light power for $\lambda_{\text{cut}} = 500$ nm.

Fig. 4 shows typical measured differential light intensity spectra of the bleaching light through the fiber from a xenon lamp with or without the dichroic reflector, or from a mercury lamp. While the spectrum with dichroic reflec-



Fig. 4. Differential light intensity spectra from a $\emptyset 0.6$ mm optical fiber are shown as a function of wavelength for a 150 W xenon lamp with and without dichroic reflector (dots and dashes) and a 200 W mercury lamp. The bleaching light power Fig.s are integrated up to 500 nm.

tor shows a cutoff at long-wave-end caused by the bandpass of the reflector, all spectra have a similar cutoff at short-wave-end caused by the bandpass of the optical fiber. Their P (500 nm) was determined to be 0.72, 1.6 and 4.3 mW respectively for these three distributions. The bleaching light power without the dichroic reflector is about 2.2 times higher than that with the dichroic, while the bleaching power of mercury lamp is about six times higher. By adjusting, focusing the intensity of the bleaching light can be changed. In this work, intensity of xenon bleaching light was adjusted to be between 1 to 2 mW.



Fig. 5. Transmittance of optical fiber and grease after irradiation.

3.5. Experimental control

Since all apparatus, except the monitors for the photofeedback unit, R4406 anode dc current and high voltage, were placed in the radiation chamber, an experimental control on performance stability under irradiation is very important. A crucial issue is to keep a stable light intensity for the bleaching light. A careful shielding was designed to protect the photofeedback unit (Oriel 68850) and the power supply.

The optical fiber and grease used, however, were exposed under radiation. A preirradiation was carried out to set both fiber and grease to a stable operation mode. Fig. 5 shows the transmittance measured for the 2 m \emptyset 0.6 mm fiber (Oriel 77517) after 3 and 100 krad irradiation. Also shown in the figure is 1 cm thick Dow Corning 200 grease before and after 0.3 and 100 krad irradiations. In the wavelength range of interest (longer than 360 nm) both the grease and the fiber provide a stable transmittance under irradiation.

4. Results

4.1. Two color centers

A more detailed study on the behavior of color centers under a bleaching light of fixed wavelength shows that 450 nm is also effective in removing color centers in BaF₂. We also found that the bleaching process can be parametrized to a sum of two exponentials, indicating two kinds of color centers in BaF₂. Fig. 6 shows measured bleachable color center density (D) of SIC302 and SIC402 as a function of integrated energy density (It) of a 450 nm bleaching light, starting from an initial dosage of 1 (crosses) and 100 krad (diamonds). The parameters of the fit to Eq. (6),

$$D = D_1 e^{-a_1 It} + D_2 e^{-a_2 It}, (6)$$

are also listed in the figure. It is interesting to find that the annihilation constants a_1 and a_2 and the ratio of the bleachable color center densities D_1/D_2 are independent of the initial dosage, and are similar for two crystals. It is also interesting to note that the quality of S402 is better than S302, as shown in a less color center density. For all these tests, crystals were wrapped with teflon film.

Similarly, Fig. 7 shows the bleaching process by using optical fiber for crystal SIC302 after an initial dosage of 1 krad. The crystal was wrapped with aluminum foil (teflon film) and the bleaching light power was 1.6 (4.3) mW for the xenon (mercury) lamp. Although the crystal shows the same two component behavior with a slow annihilation constant about tenfold smaller than the fast one, the annihilation constants are quite different, and they are very different from the case where 450 nm light was used. This is due to different bleaching light spectra of these two light sources, as shown in Fig. 4, as well as different wrappings.

Integrated Light Density: It (J/cm²) Fig. 6. Bleachable color center density of SIC302 and SIC402 after 1 krad (crosses) and 100 krad (diamonds) irradiation are plotted as a function of integrated light density for 450 nm bleaching light. The lines are a fit to a sum of two exponentials with parameter listed in the Fig..





Integrated Light Intensity: It (J)

Fig. 7. The measured bleachable color center densities of SIC302 are plotted as a function of the integrated bleaching light energy from a \emptyset 0.6 mm optical fiber for light sources of a 150 W xenon lamp and a 200 W mercury lamp. The lines are a fit to a sum of two exponentials with parameter listed in the Fig..

With a stronger UV component, the mercury lamp is more effective in bleaching the color centers in BaF_2 , and this is enhanced by the teflon wrapping.

This two color center behavior can be understood better by looking the spectra of total color center density (1/LAL)and the bleachable color center density (D) as a function of photon energy. Fig. 8 shows these spectra for crystal SIC402, starting from 100 krad until fully bleached by 450 nm light. The dashed vertical line in these figures indicate the location of 220 nm where BaF_2 's fast component resides. It is clear that the bleachable color center density at 220 nm can be decomposed to two components.



Fig. 8. The total color center density (1/LAL) and bleachable color center density (D) are plotted as a function of photon energy for crystal S402, starting from 100 krad (the highest curves in both plots), under 450 nm light bleachable. The lowest curve of the top plot is the unbleachable color center density. The six curves in the bottom plot are the difference between the top six cures and the lowest one in the top plot. The vertical dash line indicates the 220 nm light.

4.2. Self-bleaching

Since the BaF_2 scintillation light is mainly in UV range, it is conceivable that the scintillation light itself would also provide bleaching, i.e. the irradiation process itself involves both creation and annihilation. This effect would be clearly shown in the damage constant *b* determined from the irradiation measurements if different wrapping materials were used. To test this effect, two irradiations were carried out for crystal SIC301. The first one was done with teflon film wrapping, and the second with the aluminum foil. Fig. 9 shows the bleachable color center density as a function of accumulated dosage for these two cases. It is clear that the higher UV reflection of the teflon film produced a threefold smaller damage constant (*b*), and a lower total color center density, since the later itself is a result of an equilibrium.

Additional irradiation tests were carried out for all four crystals at a fixed dose rate of 123 rad/h. All crystals were wrapped with aluminum foil. The result, as shown in Fig. 10, indicates that crystals produced in a recent batch (SIC401 and 402) has less bleachable color center density than the older ones (SIC301 and 302), but the color center creation speed (b) is compatible.

4.3. Bleaching in situ

Tests were carried out to determine the light attenuation length of BaF_2 crystals which was under a dose rate of



Fig. 9. The bleachable color center density of SIC301 is plotted as a function of integrated dosage received for two different wrappings. The lines are a fit to an exponential with parameter listed in the Fig..



Fig. 10. The measured bleachable color center density of four crystals is plotted as a function of the integrated dosage received. All crystals were wrapped with aluminum foil. The dose rate was 123 rad/h. The lines are a fit to an exponential with parameter listed in the Fig..

Table 3 Transmittance (T), Light attenuation length (LAL) and bleachable color center density (D) measured for two BaF_2 samples under 130 rad/h and bleaching light

Run	time	Ι	R	Т	LAL	D
(no.)	[h]	[mW]	[rad/h]	[%]	[cm]	[1/m]
Test sa	mple: Sl	C302				
_	_		_	81.8	260	0.0
1	4.5	1.9	130	78.7	186	0.15
2	3.0	1.9	130	78.0	175	0.19
3	3.1	1.1	130	77.1	162	0.23
4	3.8	1.1	130	76.7	156	0.26
Refere	nce sam	ole: SIC30)1			
_		-	-	83.2	317	0.0
1	4.5	0.0	130	71.1	106	0.63
2	3.0	0.0	130	67.7	88	0.82
3	3.1	0.0	130	66.8	84	0.88
4	3.8	0.0	130	65.0	77	0.98

more than 100 rad/h and, at the same time, was illuminated by a bleaching light from fiber. All samples were wrapped with aluminum foil, and the setup used is shown in Fig. 1.

Four runs of a total of 14.4 h were carried out for two BaF_2 crystals. All crystals were wrapped with aluminum foil. The dose rate was set to be 130 rad/h and the intensity of the bleaching light from the xenon lamp through the fiber was adjusted between 1 to 2 mW. Table

3 lists the transmittance at 220 nm (T), corresponding bleachable color center density (D) and light attenuation length (LAL) before and after each run. While the test sample (SIC302) was illuminated by the bleaching light, the reference sample (SIC301) was placed adjacent to the test sample, but without the bleaching light. Fig. 11 shows these data as a function of integrated dosage. The data measured previously under 123 rad/h without bleaching light (Fig. 10) is also plotted for comparison. The result of this test clearly shows that with a bleaching light power of 1.1 mW from the xenon lamp, the light attenuation length of a production size BaF₂ crystal can be set to more than 150 cm under 130 rad/h irradiation. Without the bleaching light, the LAL would be reduced to about 75 cm.

Another series of tests was made to study the behavior of three BaF₂ crystals under an extreme dose rate of 400 rad/h. The bleaching light power was 4.3 mW from the mercury lamp. All samples were wrapped with teflon film in these tests. A total of five runs were carried out, where the bleaching light was applied to different samples. Table 4 lists the measured light attenuation length for the test sample (LAL_{test}) and two reference samples (LAL_{t1} and LAL_{t2}). It is concluded that about 200 cm light attenuation length can be reached at 400 rad/h by using 4.3 mW bleaching power from the mercury lamp. On the other hand, the light attenuation length would be reduced to around 70 cm without the bleaching light. Fig. 12 plots the light attenuation length result.



Fig. 11. The transmittance at 220 nm and corresponding light attenuation length and bleachable color center density for test sample SIC302 and reference sample SIC301 under 130 rad/h irradiation. Both crystals were wrapped with aluminum foil. The test sample was also illuminated by xenon bleaching light through a \emptyset 0.6 mm optical fiber. The power of the bleaching is 1.9 and 1.1. mW respectively for the first and last two runs. Previously measured data under 123 rad/h without bleaching light is also shown for a comparison.

Table 4 Light attenuation length of BaF_2 crystals under about 400 rad/h and with 4.3 mW bleaching light

Run (no.)	time [h]	<i>R</i> [rad/h]	LAL _{test} [cm]	LAL _{r1} [cm]	LAL _{r2} [cm]
1	0.5	400	231 (301)	86 (302)	89 (402)
2	3.0	370	183 (302)	97 (301)	75 (402)
3	3.0	370	186 (302)	92 (301)	74 (402)
4	2.0	370	226 (301)	85 (302)	76 (402)
5	3.0	400	212 (402)	67 (301)	67 (302)

Since a typical UV lamp provides a light spot with a few cm diameter, a lamp can drive a fiber bundle with hundreds fibers. The in situ bleaching setup thus is similar to the L3 xenon monitor system for the BGO crystal calorimeter [12].

4.4. Verification of color center dynamics

It is interesting to verify the color center dynamics model proposed in Ref. [1]. As discussed in Section 4.1, there are two color centers responsible for damage at 220 nm. We also know the annihilation constant, a, is a function of the bleaching light spectrum and the wrapping. Two sets of a's were determined for crystal SIC302: (1) with aluminum wrapping and xenon lamp bleaching and (2) with teflon wrapping and mercury lamp bleaching, as shown in Fig. 7. On the other hand, the damage constant (b) is affected by both the crystal wrapping and the dose rate, as discussed in Section 4.2. For crystal SIC302, the damage constant, b, was determined to be (1) 1.9 ± 0.2 krad⁻¹ under 130 rad/h with an aluminum wrapping, as



Fig. 12. The light attenuation length at 220 nm under 400 rad/h irradiation in various tests are plotted for three BaF_2 samples with (diamonds) and without (circles) 4.3 mW bleaching light from a mercury lamp through a $\emptyset 0.6$ mm optical fiber. All crystals were wrapped with teflon film.



Fig. 13. The measured (points) and calculated (solid line) bleachable color center densities are shown for two series of tests, where crystal SIC302 wrapped with aluminum foil (teflon film) was illuminated by 1 to 2 (4.3) mW bleaching light from a xenon (mercury) lamp through a fiber, and was under 130 (370) rad/h irradiation at the same time. The calculation was carrie out by using Equation 8, assuming two color centers.

shown in Fig. 10, and (2) $1.30 \pm 0.14 \text{ krad}^{-1}$ under 370 rad/h with a tefton wrapping.

By treating two color centers independently, we have time-dependent color center density solution

$$D = \sum_{i=1}^{2} \left\{ D_i^0 e^{-(a_i I + bR)t} + \frac{bR D_i^{\text{all}}}{a_i I + bR} \left[1 - e^{-(a_i I + bR)t} \right] \right\},$$
(7)

where D_i^0 is the initial value of the *i*th bleachable color center density. For each value of *I* and *R*, the equilibrium is established at

$$D_{\rm w} = \sum_{i=1}^{2} \frac{bRD_i^{\rm all}}{a_i I + bR}.$$
 (8)

Fig. 13 shows the measured (points) and calculated (solid line) bleachable color center density for two tests, where crystal SIC302 was with aluminum (teflon) wrapping, illuminated by 1 to 2 (4.3) mW bleaching light from a xenon (mercury) lamp through a fiber, and under 130 (370) rad/h irradiation. The calculation was done by using Eq. (7), assuming two color centers. The calculated color center density agrees well with the measured data, indicating that the color center dynamics described in Section 2 indeed describes the behavior of the dynamics of radiation damage of barium fluoride crystals.

4.5. Optical bleaching for CeF₃

It is interesting to note that optical bleaching described in this report can also be used to maintain light attenuation length for other crystals to be used in high radiation environment. Fig. 14 shows the recovery of transmittance



Fig. 14. The recovery of transmittance (top) and radiation-induced absorption coefficient (bottom) under optical bleaching with different wave lengths (700, 500, 400, 320 and 300 nm) for a 15 cm long CeF₃ crystal 30 days after 1 Mrad irradiation.

(top) and radiation-induced absorption coefficient (bottom) under optical bleaching with different wavelengths (700, 500, 400, 320 and 300 nm) for a 15 cm long CeF₃ crystal 30 days after 1 Mrad irradiation. This crystal is one of the recent CeF₃ crystals produced by SIC. It is clear that light of shorter than 400 nm is more effective in removing color centers. We also notice that the quality of CeF₃ crystals is yet to be improved, so that adequate light attenuation length can be reached with optical bleaching in situ.

5. Summary

The main results of this R&D can be summarized below.

- Two types of color centers were found in BaF_2 crystals which are bleached at different speed.
- Self-bleaching by scintillation light changes damage level of a crystal if wrapping is different, and thus changes the equilibrium point in situ.

- Newer crystals (SIC401 and 402) have less bleachable color center density than older ones (SIC301 and 302), but the creation speed is compatible.
- The light attenuation length of current production BaF₂ crystals can be set to around 200 (160) cm under 400 (130) rad/h by using 4.3 (1.1) mW bleaching light from a mercury (xenon) lamp through a Ø0.6 mm fiber.
- The color center dynamics proposed in our previous publication [1] describes the experimental data very well.
- The technical approach described in this report can also be used to maintain light attenuation length for other crystals to be used in high radiation environment.

Acknowledgements

We would like to thank Prof. D.S. Yan and Z.W. Yin for providing BaF_2 and CeF_3 samples for this work. Many inspiring and interesting discussions with Drs. P. LeCoq, P.J. Li, S. Majewski, M. Schneegans, P. Schotanus, R. Sparow, C. Woody, C. Wuest, D.S. Yan and Z.W. Yin, are also acknowledged. The work presented in this report is supported in part by US Department of Energy Grant No. DE-FG03-92-ER40701.

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