Recovery of Radiation Damage in Lead-Glass by Thermal Annealing

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1 Abstract

A fundamental goal of nuclear science is an understanding of the basic constituents of matter, quarks and gluons, and how they interact. A key ingredient in this understanding is a description of the internal structure of the nucleon. The elastic electric form factor (GEp) experiment aims to measure properties that directly relate to the charge and current distributions of the proton. Electrons from the experiment are detected by an electromagnetic (EM) calorimeter based on lead-glass blocks. High-energy electrons that interact with the lead nuclei create a shower of electrons and photons. The electrons and positrons travel at very high velocity within the medium of lead-glass and produce Cherenkov photons. Due to its density and transparency, lead-glass is a good material to be used in the electron calorimeter (Ecal). The material, however, suffers from radiation damage and a loss of transparency during operation. The transparency can be recovered through thermal annealing, but the timescale and affect of temperature on the annealing process in lead-glass need further investigation before implementation in future GEp experiments. A transparency measurement was conducted by shining a low power laser through a block of damaged lead-glass as the block is heated and its transmission monitored. Additionally, blocks of lead-glass were placed in an oven as temperature-time profile as well as the reduction of damage were recorded. From these temperature profiles, an approximation for the characteristic annealing time, t, as a function of temperature was extracted. A thorough understanding of thermal annealing in lead glass would allow GEp experiments to maintain high energy resolution and contribute to the success of future experiments.

Contents

2 Introduction

2.1 Calorimetry

GEp will be made up of several detector components, many of which will be calorimeters - devices that measure the energy of an incoming particle. The experiment will include a hadron calorimeter to be place in post-collision path of the proton (the proton arm) and an electromagnetic calorimeter will be part of the electron arm. The electromagnetic calorimeter will be homogenous, in which the entire volume of the calorimeter will be sensitive, as opposed to a sampling calorimeter with alternating layers of absobers and sensitive material. The calorimeter works be measuring the energy deposited in a carefully selected material via an electromagnetic shower. The incoming electron within the calorimeter's medium will travel faster than the speed of light and create Cherenkov photons. These photons will then deposite energy through the production of an e^+/e^- pair, interacting with the nuclei of the medium to conserve momentum. These charged particles will, in turn, create more photons and in a continuous "shower" of photons and e^+/e^- particles that ends when the electron reaches it's critical energy within the chosen medium.

2.2 Use of Lead Glass in ECal

A homogeneous electromagnetic calorimeter (ECal) will be constructed for the GEp experiment as a part of the electron arm. Lead-glass is a useful material to be used in ECal due to its transparency, density, and relatively low cost. Transparency and high density of lead glass are important for a homogeneous calorimeter because it allows the glass to act as both the absorber and the active medium in which an electromagnetic shower can occur. Its low costs allows for a higher active volume for Ecal.

2.3 Radiation Damage

The transparency of lead-glass is compromised when it is exposed to high levels of radiation, radiation that has sufficient energy to ionize the atoms in the lead-glass. Due to a very low electrical conductance, the ions of the lead-glass and the now free electrons remain separated. These newly-formed ions change several properties of the lead-glass and are much more likely to interact with photons. After sufficient damage, the lead-glass suffers a significant decrease in transparency, becoming tinted. (See figure 1). As the absorption of the lead-glass increases, the intensity of transmitted light decreases and the energy resolution of the calorimeter as a whole decreases. Radiation damage in lead-glass resulted in the energy resolution of a calorimeter in a previous GEp experiment to go from $\sigma_E/E = 12\%$ to 28% for E equal 1GeV [1]. With the radiation expected to be much larger in future GEp runs, a method to recover radiation damage is needed.

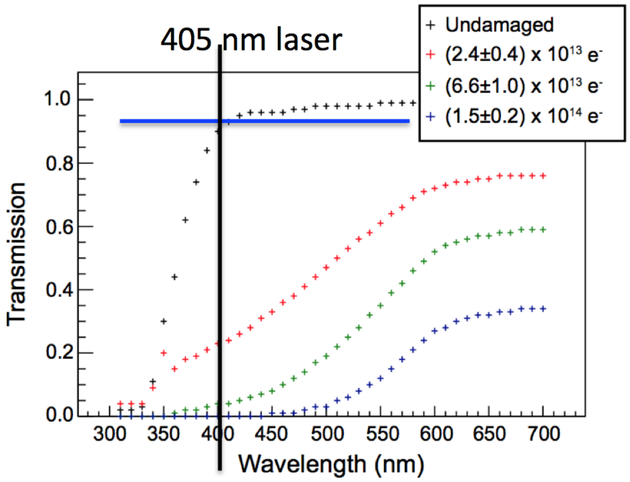

Figure 1: Transmission of lead-glass exposed to various levels of radiation vs.wavelength[2].

An important characteristic of the radiation damage is that it is not homogeneous along the lead-glass. The front of the lead-glass that is initially exposed to the incoming radiation is usually the most affected. Figure 2 shows a much lower transmission at the front of the bar.

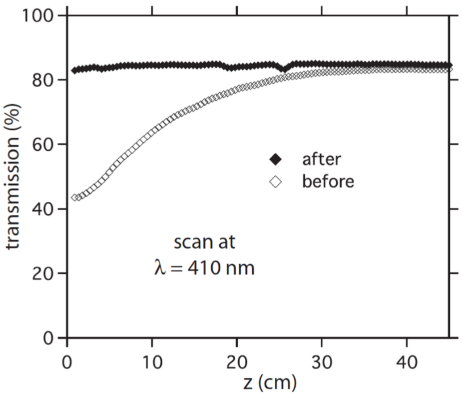

Figure 2: Transmission along 4 cm thick lead-glass bar before and after annealing at 260 °C for 12 hours[3].

2.4 Calorimeter Parameters for Future GEp Run

The lead-glass calorimeter to be used in the GEp experiment will need to detect 4-5 GeV electrons with an energy resolution of at least 10% for 3.5 GeV electrons and a spatial resolution of 6-8mm. The proposed calorimeter will be made up of 2000 lead-glass blocks in a rough ”C” configuration and an aspect ratio 1:4. The radiation damage of lead-glass and the resulting loss in transparency pose a problem because they contribute to a loss in energy resolution. The calorimeter will use a continual bake out scheme in which the apparatus containing the lead-glass blocks will be heated at an ideal annealing temperature to offset radiation damage.

2.5 Annealing

A known method for recovering lead-glass from radiation damage is thermal annealing. Upon heating, the electrical conductivity of the lead-glass also increases. This allows free electrons to neutralize the ions in the lead-glass blocks and restores the original properties of the material. Figure 2 shows recovery of transmission of the lead-glass bar back to roughly 84 % through the 4cm thick block, the maximum transmission expected from lead-glass of this thickness after taking into account the reflection at the boundaries of the block (see Calculation of Transmission section).

2.6 M200

The prototype to test the concept of a heated calorimeter will involve placing 1884 42 x 42 x 400 mm^3 lead-glass blocks in a custom oven heated to 200°C. At this temperature, the loss in transparency due to radiation damage is greatly reduced. The lead-glass will be stacked in 20 rows of 10, with each row shifted by half the width of a single block (21 mm). This stack of lead-glass will be held in place by surrounding blocks of stone. See Figure 1.

The back end of the lead-glass will need to be connected to photo-multiplier tubes (PMTs). The two cannot be connected directly however, as the PMTs would melt at 200°C. A 150cm-long soda-lime glass cylinder with diameter 30 mm will act a a light path connecting the two components. The light path will be connected to the lead-glass via a heater base. The opposite end of the light path will be cooled to 20°C and connect to the PMTs, and lastly dividers. This set up will be used on the first 50 blocks of the prototype.

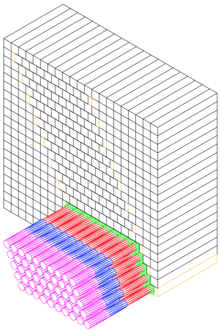

Figure 3

Figure 3 shows a model of this setup , with green as the heater bases, red as the cylindrical light path, blue as the PMTs, and magenta as the dividers. Only the first 50 blocks of lead glass from the bottom will contain this setup.

Data from this prototype, as well as results from invetigation of thermal annealing will be used in the development of ECal.

3 Calculation of Transmission

The expected transmission of light through lead-glass was calculated using the the known absorption of roughly 4 % per 4cm of lead glass and the Fresnel equations for the reflection at the boundaries of the block.

The Fresnel equation for the *transmission coefficient* for a material with refractive index n_t is given by

$$\frac{2n_i\cos(\theta_i)}{n_i\cos(\theta_i)+n_t\cos_t(\theta_t)}$$

where n_i is the refractive index of the material the light is coming from.

Our experiment involves shining light that is perpendicular to the block of lead-glass. Thus both θ_i and $\theta_t = 0$ and the equation for the *Transmittance* simplifies to:

$$T = \frac{4n_in_t}{(n_t+n_i)^2}$$

T, the Transmittance was calculated to be about 0.94 for one boundary, with n_i as the refractive index of air ≈ 1, and $n_t =$ index of refraction of lead-glass = 1.65 .

We then calculated the transmission of the light through the lead glass itself. for 4 cm of lead-glass, the transmittance is 0.96 .

With a block of length z, and incorporating both boundaries, the total transmission of a block of lead-glass is

$$T_{total} = T_{boundaries}\cdot T_{length} \approx 0.94^2\cdot 0.96^{\frac{z}{4}} \quad (1)$$

For a 35cm-long block of lead-glass, we calculated to the transmission to be 0.618 .

Very close to our experimental value of 0.607 .

4 Propagation of light in Lead-Glass

The signal expected from our detector will be proportional to the energy of the incident electrons and the number of Cherenkov photons. S, the signal, is given by the expression under the integral:

$$S_{ideal} = \int z^{1.75}e^{-\frac{z}{6}}\frac{c}{\lambda^2}Q(\lambda)d\lambda dz \quad (2)$$

1.75 and 6 are parameters derived from the specific properties of lead-glass and z in cm.

The expression $z^{1.75}e^{-\frac{z}{6}}\cdot\frac{c}{\lambda^2}$ represents the longitudinal distribution of the shower produced by the electromagnetic shower in the lead glass with $\frac{C}{\lambda^2}$ relating to spectrum of Cherenkov photons and $Q(\lambda)$ the quantum efficiency of the phototube.

With the transmission of light being related to the quantum efficiency, or probability of a photon-electron conversion, the absorbed signal can also be written as

$$S_{real} = \int F_1(z)T(\lambda,z)dzd\lambda \quad (3)$$

The transmission of light through a 40cm long block of lead glass, the typical length of block to be used, is given by:

$$T(\lambda,z) = exp[-\int_z^{40}\frac{dy}{a(y,\lambda)}] \quad (4)$$

The above equation shows that the transmission has a dependence on the absorption length of the material, $a(\lambda,z)$, as well as the location along the block. The dependence on position is due to a non-uniform distribution of radiation damage along the block (recall Figure 2), as well as the distance the light must travel through the block.

The absorption length, $\tilde{a}(y,\lambda)$ refers to the distance within the lead glass at which the probability of a photon being absorbed within the material is $1-e^{-1}$.

The absorption length of undamaged or fully annealed, 4cm-thick lead-glass bar can be expressed as follows:

$$\tilde{a}(\lambda) = -\frac{4}{\ln T_4} \quad (5)$$

The inverse of the absorption length of a damaged lead-glass block can then be written as the inverse sum of undamaged and damaged lead-glass absorption lengths:

$$\frac{1}{a(z,\lambda)} = \frac{1}{\tilde{a}(\lambda)} + \frac{1}{b(z,\lambda)} \quad (6)$$

The relationship between the absorption length of undamaged lead-glass, $\tilde{a}(\lambda)$, and Transmission was outlined in equation (6). $a(z,\lambda)$, however, can also be determined from the transmission. For a damaged block of lead-glass, the length at which the intensity falls to $1-e^{-1}$ of it's original intensity is the absorption length for that sample of lead-glass. With $a(z,\lambda)$ and $\tilde{a}(\lambda)$ determined from transmission measurements, we can also determine parameter $b(z,\lambda)$ from equation (7).

The goal of annealing is the obtain a longer absoprtion length, $b(z,\lambda)$.

The cross section for the lead-glass, or the probability of light interacting with our material, can be written as the sum of the cross section of undamaged lead-glass and the product of the cross section of damaged lead-glass and the concentration of damage along the block:

$$\sigma = \sigma_N + \sigma_c \cdot C(z) \tag{7}$$

5 Transmission Monitoring During Annealing

5.1 Setup

Annealing of lead-glass to recover from radiation damage is a well known procedure. The purpose of this experiment is to gain a better understanding of the time scale at which this damage is recovered.

Annealing Oven.png

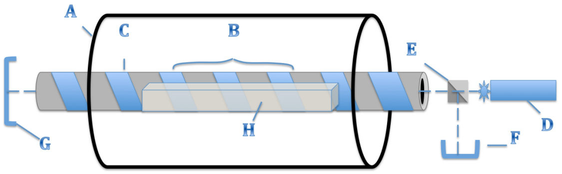

Figure 4:
A .Outer Shell
B. Inner Tube
C. Heating Coil
D. 405nm Laser
E. Beam Splitter
F. Photometer
G. Photometer
H. Lead-Glass

A 40 cm-long damaged lead-glass block was placed in a custom oven that was quickly heated to annealing temperature. The block is placed within a tube with several metal coils surrounding it to heat the lead-glass. Outside of this tube, 10 cm of glass wool surrounds the block followed by 15 cm of air.

A beam splitter is attached to a low-powered 405 nm pocket laser. Directly underneath the beam splitter is a small photometer that receives a fraction of the light. On the opposite end of the oven is another photometer. When the light is turned on both photometers are read in microamps from two separate Ammeters with intensity of light is read as the current from the photometer.

The oven was powered through a variable autotransformer. We controlled the temperature of the oven by adjusting the voltage on the transformer. To ensure the beam hit the center of the photometer opposite the laser, we created a small screen that was placed over the opening of the photometer. This allowed us to see the outline of the opening on the photometer as the beam shined through.

End caps and convection shields were constructed out of aluminum. The end caps were installed in order to prevent air from entering the tube for faster heating. The purpose of the convetion shields was to restrict air flow within the tube and make the heating more uniform along the tube.

Lastly, Tedlar sheets were placed over both photometers. This reduced the dark current on either photometer to 0-3 nA.

5.2 Procedure

The autotransformer was turned to 55 volts in order to reach an annealing temperature of 250°C quickly. As it approached annealing temperature, the voltage was lowered. When the glass was sufficiently heated for a specific trial, the oven was turned off and the glass set to cool. Throughout the entire process the transmission was regularly monitored. The laser would be turned on and the ratio of two ammeters connected to the to photometers was recorded.

6 Measurement of Temperature Profiles

For additional studies, blocks of lead glass were placed in a larger, closed oven. A thermal couple was attatched direclty of the damaged portion of the lead-glass with the entire block insulated by fiber-glass. The oven was programmed to ’soak’ at a specific temperature for a set amount of time. To get to the ’soaking’ temperatures, however, there were initial and ending periods of ’ramping’ during which the oven heated or cooled to the appropriate temperature. An effective time of annealing was calculated to be the interval of time during which the temperature was within 30°of maximum. The average temperature over this period of time was calculated and considered to be the effective temperature for the specific trial.

The temperature-time profile of the glass was taken by recording the temperature of the block at regular time intervals. The transmission (expressed as a ratio of light intensity, R) was recorded for each block before and after their session in the oven. This detail proved to be very important, as the measurement would have been slightly distorted had we not let the glass completely cool before hand. From our measurement of the temperature profile, we integrated temperature over time, and had several ratios relating to the transmission before and after annealing.

7 Transmission as a Function of Annealing Time and Temperature

The transmission of an entire block of lead glass can be expressed in terms of the absorption length

$$Tr = exp[-\int_0^{45} \frac{dx}{\lambda(x)}] \tag{8}$$

Similar to equation (4), but with $\lambda(x)$ as the absorption length of the block, and the limits of integration being 0-45 to represent a 45cm block.

Recalling equation (6), we can express the the absorption length of the block as a combination of the absorption length for damaged and undamaged lead glass:

$$\frac{1}{\lambda(x)} = \frac{1}{\tilde{\lambda(x)}} + \frac{1}{\lambda_{damaged}} \tag{9}$$

The absorption length can then be rewritten in terms of the concentration of damage, and the characteristic annealing time, τ:

$$\frac{1}{\lambda_{damaged}} = C = C_0 \cdot exp[^{-}\int \frac{dt}{\tau}] \tag{10}$$

The characteristic annealing time, after which the concentration of absorbing centers falls by a factor of e, is expected to behave like the electrical conductivity of lead-glass as a function of temperature:

$$K \propto e^{\Delta\beta/kT}$$

with T as temperature in Kelvin. We then approximated τ as

$$\tau = \tau_0 \cdot e^{-b/T} \tag{11}$$

Substituting equation 11 into 10 yeilds:

$$C = C_0 \cdot exp[-\int \frac{dt}{\tau_0} e^{b/T}] = C_0 \cdot exp[-J(b)/\tau_0] \tag{12}$$

We can then make the appropriate substitutions to express transmission as:

$$Tr = exp[-\int (\frac{1}{\tilde{\lambda}(x)} + \frac{1}{\lambda_{damaged}})dt$$

$$Tr = exp\{-\int \frac{1}{\tilde{\lambda}(x)}dt - \int_0^{45} C_0 \cdot exp[-J(b)/\tau_0]dt\}$$

And further simplify to:

$$Tr = exp[-(a_1 + a_2 \cdot e^{-J(b)/\tau_0})] \tag{13}$$

7.1 Calculation of a1 and a2

By shining the laser directly through the block, we can measure the ratio of the light intesity through the block and through air, $\frac{I}{I_0}$, denoted as R - from which transmission is directley related (obtained from R by simply correcting for reflection at the boundaries of the glass). Several of these ratios were taken before and after the glass was placed in the closed oven.

$$\ln R = -a_1 - a_2 \cdot e^{-J(b)/\tau_0} \tag{14}$$

Because the right hand side of the sum is dependent on the concentration of ions, when measuring a completely clean block of lead-glass, we can directly extract a_1 as $a_2 = 0$

We can then extract the a_2 term by finding R for an unnanealed damaged block. In this instance, the exponent = e^0 and the a_2 term is non-zero.

With the a_1 and a_2 terms known, we can begin to calculate b

7.2 Damage Reduction Factor

The damage reduction factor can be determined by finding the ratio of the contribution of damage to the light absorption in lead-glass before and after annelaing:

$$DR = \frac{-a_2}{\ln R_{after} + a_1} \tag{15}$$

7.3 calculation of b

By creating another ratio from the transmission a glass block before and after annealing, we obtain

$$\ln(\frac{\ln R_1}{\ln R_2}) = -J(b)/\tau_0 \tag{16}$$

We can then compare the ratio of two blocks, blocks α and β, with different annealing parameters and get an experimental value for that ratio:

$$\frac{J_\alpha(b)/\tau_0}{J_\beta(b)/\tau_0} = \frac{J_\alpha(b)}{J_\beta(b)} = \frac{\ln(\frac{\ln R_1}{\ln R_2})_\alpha}{\ln(\frac{\ln R_1}{\ln R_2})_\beta} \tag{17}$$

Using the measurements of the temperature profiles of lead glass during annealing, we can calculate the integrals $J_\alpha(b)$ and $J_\beta(b)$ and for various b values. The correct b is used when equation 17 is satisfied.

7.4 Obtaining Chracteristic Annealing Time

We aimed to approximate the time it takes for the contribution of damage to the absorption of light in lead glass to decrease by a factor of e at various temperatures, T. With equation 16, $\ln(\frac{\ln R_1}{\ln R_2}) = -J(b)/\tau_0$, and our calculated value of b, we can first obtain τ_0

with τ_0 and b now known, we can obtain τ for various annealing temperatures with equation 11:

$$\tau = \tau_0 \cdot e^{-b/T}$$

From this equation, we can also obtain a more immediately useful equation, with the annealing temperature of 200 °C (473°K), τ_{200}, as a base rather than τ_0

$$\tau = \tau_{200} \cdot e^{-b \cdot (\frac{1}{T} - \frac{1}{473})} \tag{18}$$

8 Results

8.1 Transmission Monitoring

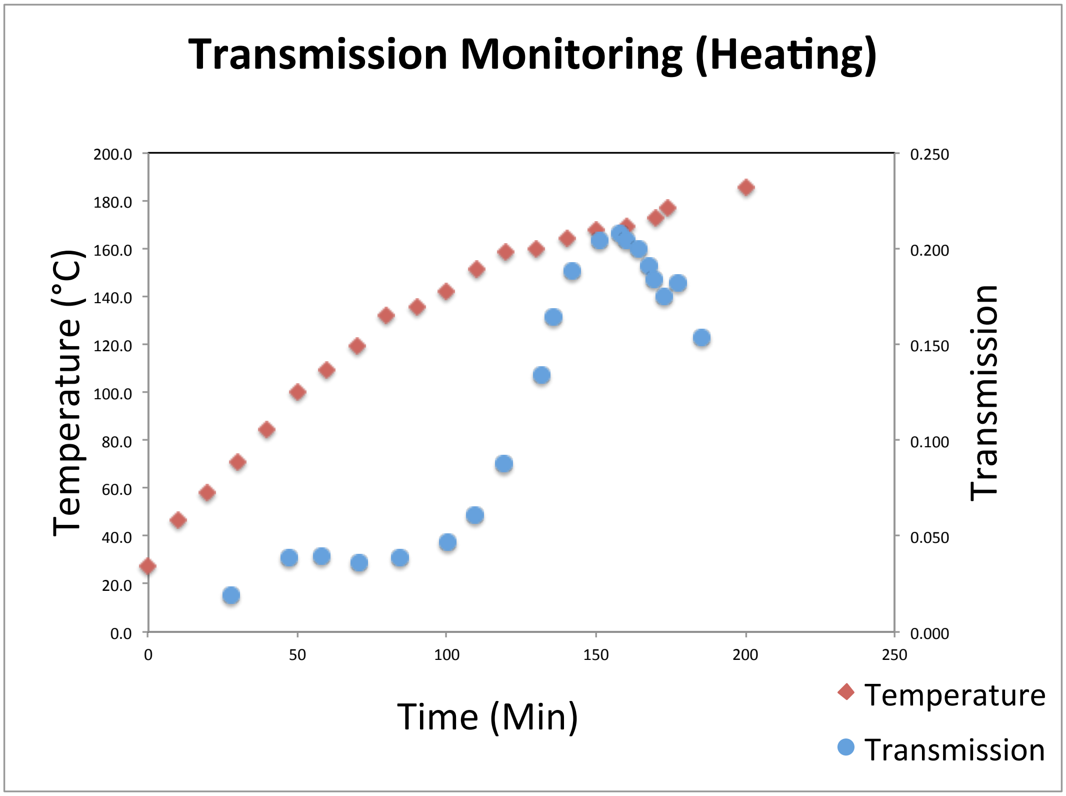

Figure 5: A 45.5 cm block of lead-glass. Its transmission is monitored during its time in the oven as it heats. The initial and rapid increase in transparency due to a reduction in the concentration of absorbing centers was expected. However, as the oven continues to heat beyond 160 °C, the transparency unexpectadly decreases.

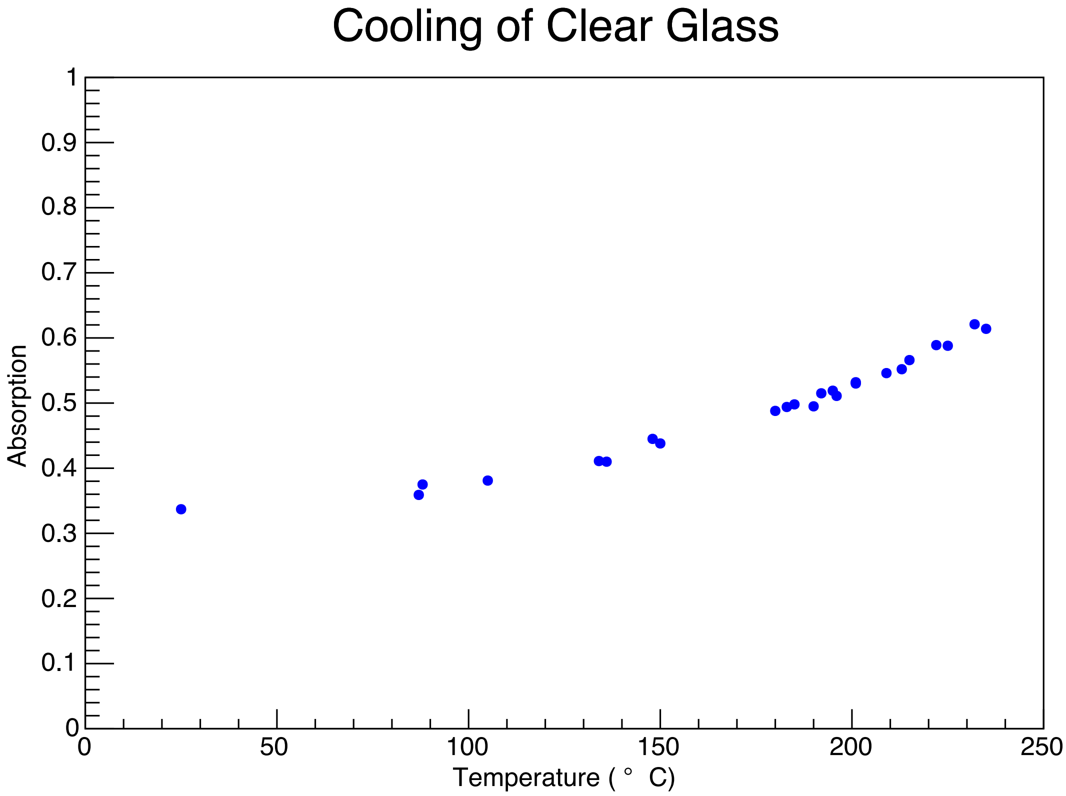

Figure 6: To verify the relationship between heat and transparency, a 45.5 cm clear lead-glass block is heated to 235 °C and set to cool as the transmission is monitrored. It was found that at 200°C, the absorption of light increases by a factor of 1.6 .

8.2 Temperature Profiles

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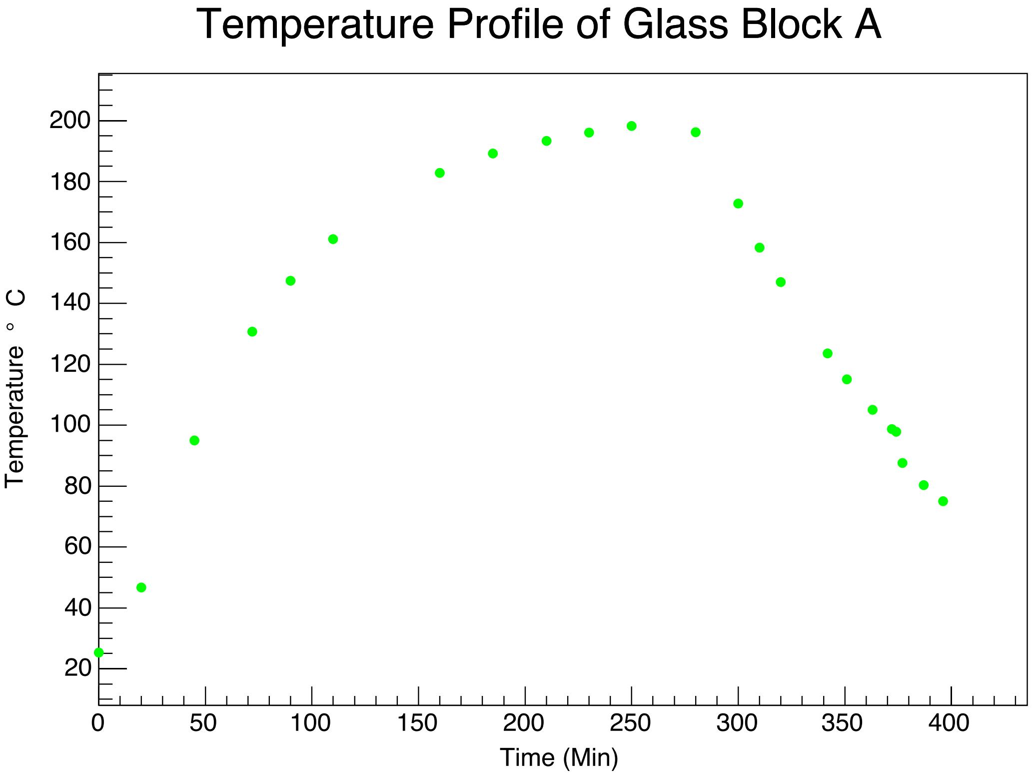

Figure 7: The oven was programmed for 4 hours at 200 °C. The effective time of annealing was 190 minutes, or the time during which the temperature was 30° within the maximum. The average temperature during this period, or the effective temperature, was 186 °C.

6 prof.pdf

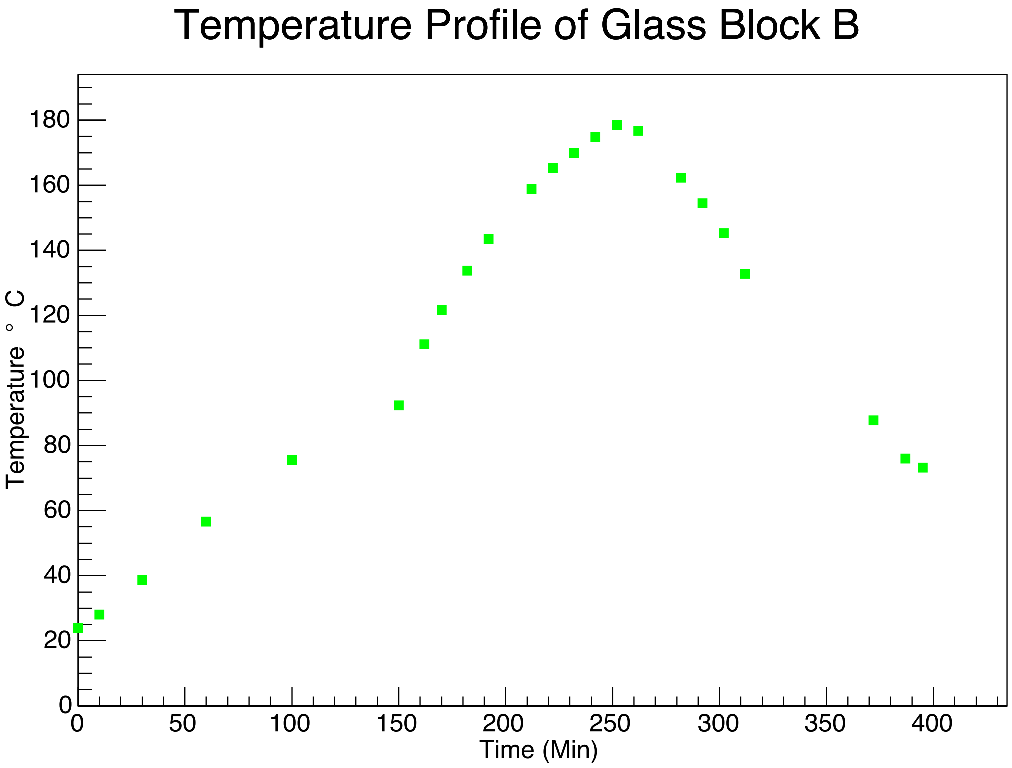

Figure 8: Block B was placed in the oven programmed for 2 hours at 200 °C. The effective time was 100 minutes with an effective temperature of 165 °C.

Profile Block 7.pdf

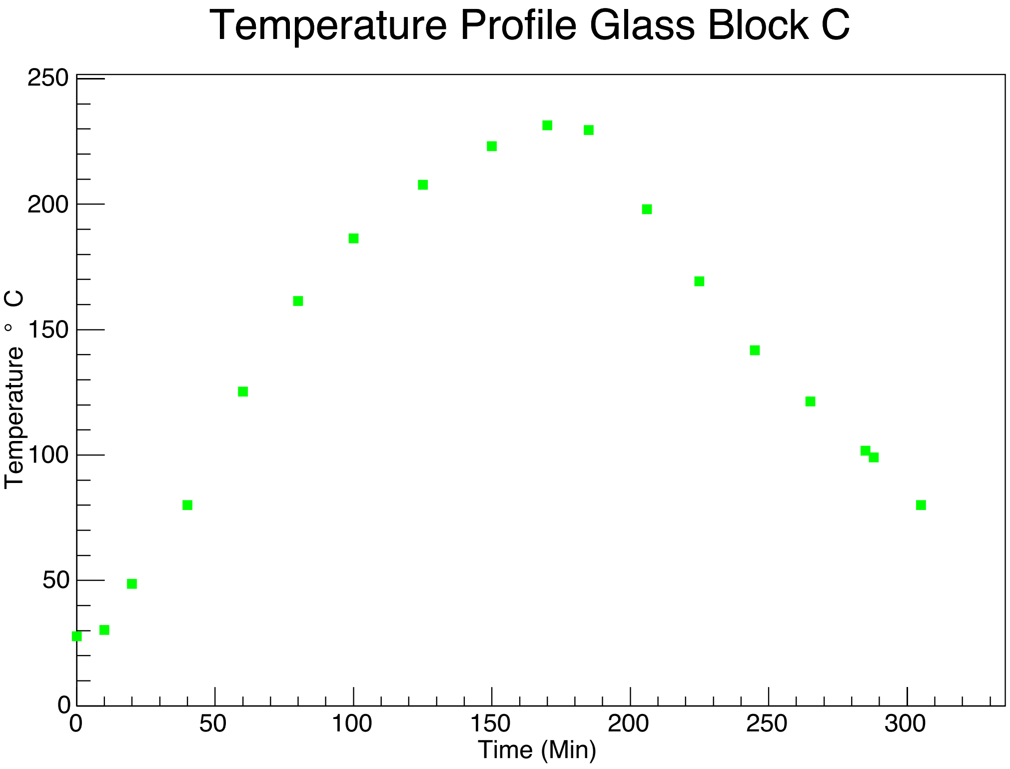

Figure 9: The thrid and final block, block C, placed in the oven set for 2 hours at 250 °C. The effective time of annelaing was 81 minutes at an effective temperature of 218 °C.

Block	T_{set}	T_{eff}	Δt	a_1	a_2	DR	τ = Δt/lnDR
A	200	186	4	0.499	2.58	11.22	1.65
B	200	165	2	0.499	2.52	3.6	1.56
C	250	218	2	0.499	1.72	2.87	1.9

Figure 10: Displayed are two temperatures: the set temperature of the oven, T_{set} and the effective annelaing temperature of the glass, T_{eff}. Δt is the set time of the oven, a_2 is the term related to the initial concetration of absorbing centers, DR is the damage reduction factor, and τ is the obtained charactersitic annealing time approximated using Δt and the natural log of the damage recover factor. It is important to acknowledge that block C had lower initial concentration of absorbing centers, and observed a longer τ than expected.

9 Conclusion

We obtained a characteristic annealing time, τ, of 1.75 hours at an effective temperature of 165 °C. The damage was reduced by a factor of 3.6 with 1.6 hours at the effective temperature. A damage reduction factor of 11.2 was obtained with 3.2 hours at an effective temperature of 186 °C. The monitoring of transmission of a clear block of lead-glass revealed that the absorption of light increased by a factor of 1.6 at 200 °C, leading to a 24% decrease in transmission. By analysing the temperature profiles for blocks A-C we found that the time in which the concentration of absrobing centers decreases by a facotr of e can be approximated by $\tau = \tau_0 \cdot e^{-b/T}$. To verify this result however, several more tests with blocks of varrying initial ion concentrations will need to conducted

10 references

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