Response of CR-39 to Heat Exposure

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Abstract

As a nuclear track detector, CR-39 (polyallyl diglycol carbonate) is used to reveal the products of fusion reactions. Positioned around the OMEGA target chamber, pieces of CR-39 absorb and are damaged by ejected particles. The plastic is then etched in NaOH solution at 80°C for 6 hours, resulting in the formation of nuclear tracks visible under a digital microscope. The etching process also reveals background noise, speculated in part to be the result of exposure to heat during shipment between suppliers and laboratories. To determine the effects of heat exposure on data analysis, pieces of CR-39 were exposed to high temperatures for various times. Heated pieces were then exposed to protons of known energies using the particle accelerator at SUNY Geneseo. Each exposed piece was etched and compared to a control. Pieces were found to oscillate between a visibly cloudy and clear appearance as a function of time exposed to high temperatures. This property was measured and quantified via reflectance and transmission of light. Furthermore, it was observed that exposure to temperatures above 100°C resulted in inaccurate data collection as etch rate and therefore track size were affected. This combination of results is an indication that current tests of the purity of CR-39 before use on OMEGA may not be sufficient.

1. Introduction

The Laboratory for Laser Energetics (LLE) is currently investigating inertial confinement[1] as a viable means of inducing and sustaining fusion reactions as an alternate source of power. Using the OMEGA 60-beam laser system fuel capsules are imploded, achieving pressures and temperatures high enough to overcome the electrostatic repulsion of nuclei to be fused. Once these extreme conditions are produced, deuterium and tritium atoms are able to undergo a reaction in which a helium atom and a high energy neutron are formed as products.[2]

For research to progress it is imperative that an accurate record of the products of these implosion reactions be generated.

The nuclear track detector CR-39 is used to fulfill this purpose. A relatively inexpensive diagnostic, CR-39 is exceptionally sensitive to ejected protons and other larger particles produced during implosion.[3] Through the use of filters, CR-39 can also be prepared to register particles of a broad range of energies. The chemical composition of this plastic is homogenous and isotropic allowing for the creation of an accurate calibration of observed measurements independent of detector orientation.[3]

While particles are produced with constant energies, the energies with which these particles reach detectors vary. Data collected from CR-39 diagnostics can be used to quantify the amount of energy lost between particle production and absorption by the detector. This information can then be used to calculate the fuel areal density[4] as almost all energy loss can be attributed to collisions between ejected particles and particles of the unfused fuel between core and detector as reactions are initiated in vacuum environments, minimizing collisions with unrelated particles. The fuel areal density can further be used to determine how much fuel was successfully fused and how much fuel did not react. Thus CR-39 detectors provide a means by which to quantify the success of individual reactions.

Before the initiation of reactions, pieces of CR-39 are positioned around the periphery of the target chamber. This chamber is then evacuated and a sequence of laser pulses is fired at a target in the center of the chamber. During a burn interval of about 150 ps CR-39 diagnostics are bombarded by the high energy products of implosion reactions.[4] These particles, including protons, alpha particles, and deuterium and tritium atoms, penetrate the surface of detectors, breaking bonds in the plastic along tracks of depths proportional to the initial kinetic energies of

ejected particles. These tracks generally range in diameter from 3 to 10 nm,[3] a size too small to be made visible with a digital microscope.

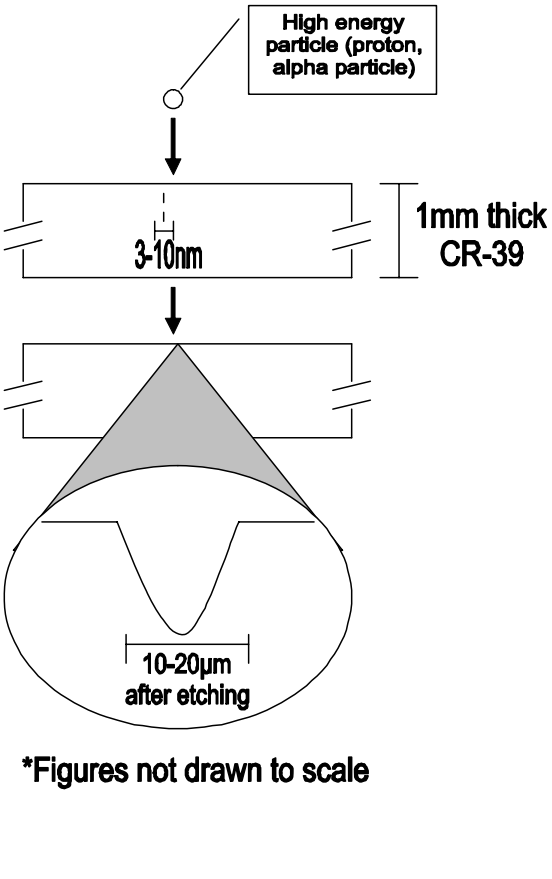


Figure 1: Damage tracks in CR-39 detectors

Detectors are removed from the target chamber after each reaction and for this experiment immersed in six-molar NaOH solution for six hours. This solution is maintained at a constant temperature of 80°C throughout the etching process by resting solution containers in an 80°C water bath. Throughout the six hours of etching, undamaged plastic is removed at a bulk etch rate. The plastic surrounding tracks is removed at a higher rate, the track etch rate, thus forming conical pits with depth and diameter proportional to particle energy.[3] The process by which pits are formed is shown in Figure 1. These pits, about 10-20 μm in diameter, are visible under a digital microscope. The etched tracks can thus be examined using the Charged Particle Spectroscopy (CPS) program developed by scientists at MIT.

This program first observes eccentricity of pits to eliminate tracks of particles that did not impact the detector normal to its surface. While particles approaching the detector at a 90° angle produce circular tracks, the particles approaching from other angles have tracks of greater eccentricity. Contrast is then evaluated with darker pits being those caused by higher energy

particles and lighter pits resulting from the impact of lower energy particles. Finally, pit diameters are measured, larger pits indicating lower energies and smaller pits indicating higher energies, to determine the energy spectra of particles ejected during implosion. It is important to note, however, that measured pit diameters correlate to different energies depending on the mass of the particle by which such pits were created. A sample image of CR-39 exposed to one type of particle is shown in Figure 2.

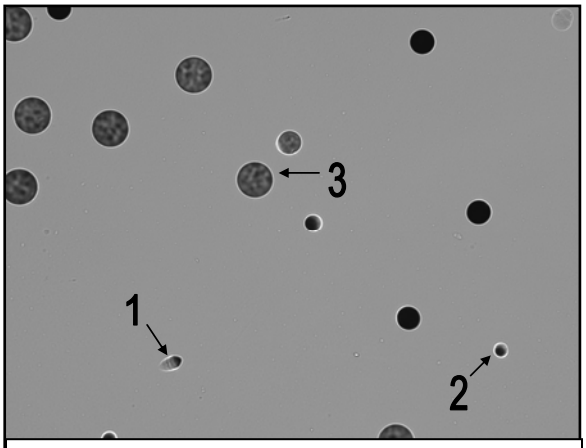


Figure 2: Digital microscope image
Pit 1 would be eliminated in the CPS system because of its high eccentricity. Pit 2 would be identified as resulting from the impact of a high energy particle while pit 3 would be identified as being created by a low energy particle.

With differences in diameter of less than 1μm significantly changing measured energy, small errors can compromise collected data. While CR-39 is ideally stored in a freezer at about 0°C, it can be exposed to temperatures well above 40°C during shipment despite efforts to maintain a low temperature environment including shipping in coolers and with ice. Thus it became necessary to investigate the effects of heat exposure during transportation from the diagnostic manufacturer: Track Analysis Systems Ltd. (TASL) in England, and between LLE and Massachusetts Institute of Technology (MIT).

2. Pit Diameter Change

For this experiment, pieces of CR-39 were stored in a freezer at 0°C prior to use. The frames were etched to reveal any visible impurities. Samples near compromised frame areas were removed.

The remaining samples were exposed to temperatures ranging from 100°C to 150°C ±1°C for 2 to 24 hours ±1 minute. To accomplish this, each piece was set in a preheated

autoclave for the designated time. No time was allowed for gradually warming. Each piece was then taken from the autoclave directly into a room at about 25°C and left to cool for at least 24 hours. The samples were then taken to SUNY Geneseo where each was individually exposed to accelerated protons with energies of 1.000, 1.500, and 3.000 MeV±0.022 MeV.

Because of the high density of the particle beam, it was necessary to scatter particles off a gold foil target instead of directly exposing the CR-39. As the mass of the nucleus of a gold atom is very high in comparison to the mass of a proton, a relatively elastic collision can be assumed to occur between the accelerated proton and gold foil. Samples were exposed in a vacuum environment to avoid energy loss in collisions with other molecules. Thus the measured beam energy can be assumed equal to the energy of particles impacting the CR-39 surface within a small error. Each piece of CR-39 was secured in a spring loaded holder set at a scattering angle of 58° to the gold foil target. The surface of the CR-39, 142.7 mm from the foil target, was protected by a 0.0125 mm thick aluminum filter. A Rutherford backscattering grazing (RBG) detector was set at an angle of -58° to the foil target. This detector was measured to be 63.48 mm from the foil target.

In order to regulate the density of particles hitting the CR-39 detector, ideally 5000 particles/cm^2, it was necessary to calculate the corresponding number of particles registered in the RBG detector. For this calculation the exposed area of the RBG detector was measured to be 0.07800 cm^2; the exposed area of the CR-39 detector was measured to be 10.88 cm^2. From this data the ideal number of particles impacting the CR-39 surface was determined to be 5.440×10^4 particles. The solid angle of each detector was then calculated by dividing the detector's surface area by the square of its distance from the foil target. Finally a proportion was set up in which the number of particles impacting the CR-39 detector divided by the CR-39 solid angle was set equal

to the number of particles impacting the RBG detector divided by the RBG solid angle. This equation was solved giving 1.971×10^3 particles as the desired number of particles to be registered in the RBG detector. It was not necessary however to register this exact number of particles as the main purpose of density regulation was to avoid the overlapping of pits after etching.

After being exposed samples were returned to LLE, etched, and scanned using the CPS program. Measured pit diameters for each of the three energies were then compared to expected diameters according to the calibration curve developed at MIT. Pit diameters measured from the control piece were found to fit this calibration. Diameters of pits created by particles of 1.022, 1.522, and 3.021 MeV particles correlated to energies of 0.923, 1.472, and 3.240 MeV respectively with errors of 9.69%, 3.29%, and 6.82%.

Heating for two hours at 100°C prior to exposure introduced large errors. Pit diameters changed drastically such that the diameters of pits created by particles with energies of 1.022 MeV were too large to fall within the range of the calibration curve for CR-39, indicating impact by particles of energies too small to be registered by a CR-39 detector. The measured diameters of pits created by particles of energies 1.522 and 3.021 MeV correlated to energies of 1.056 and 4.743 MeV respectively. Thus heating detectors at 100°C for two hours prior to bombardment with energetic particles introduced errors of 30.6% and 57.0% in the measured diameters of pits created by 1.522 and 3.021 MeV protons.

Heating at higher temperatures prior to exposure also introduced large errors. Data from some particle impacts was lost, with only one peak appearing on a histogram of measured pit diameters (see Figure 3). As all samples were exposed to a consistent number of protons at three controlled energies, each diagram should display peaks similar to those shown in the control

sample graph. However, at temperatures of 100°C the three peaks are shifted to include tracks of larger diameters. Fewer tracks were accepted by the CPS system in these shifted peaks. At higher temperatures, only one peak, excluding that of the background noise, is measured. The central portion of this peak accounts for considerably fewer tracks than those of the control sample. The appearance of only one peak indicates exposure to particles of only one energy despite actual exposure of all samples to particles of three distinct energies.

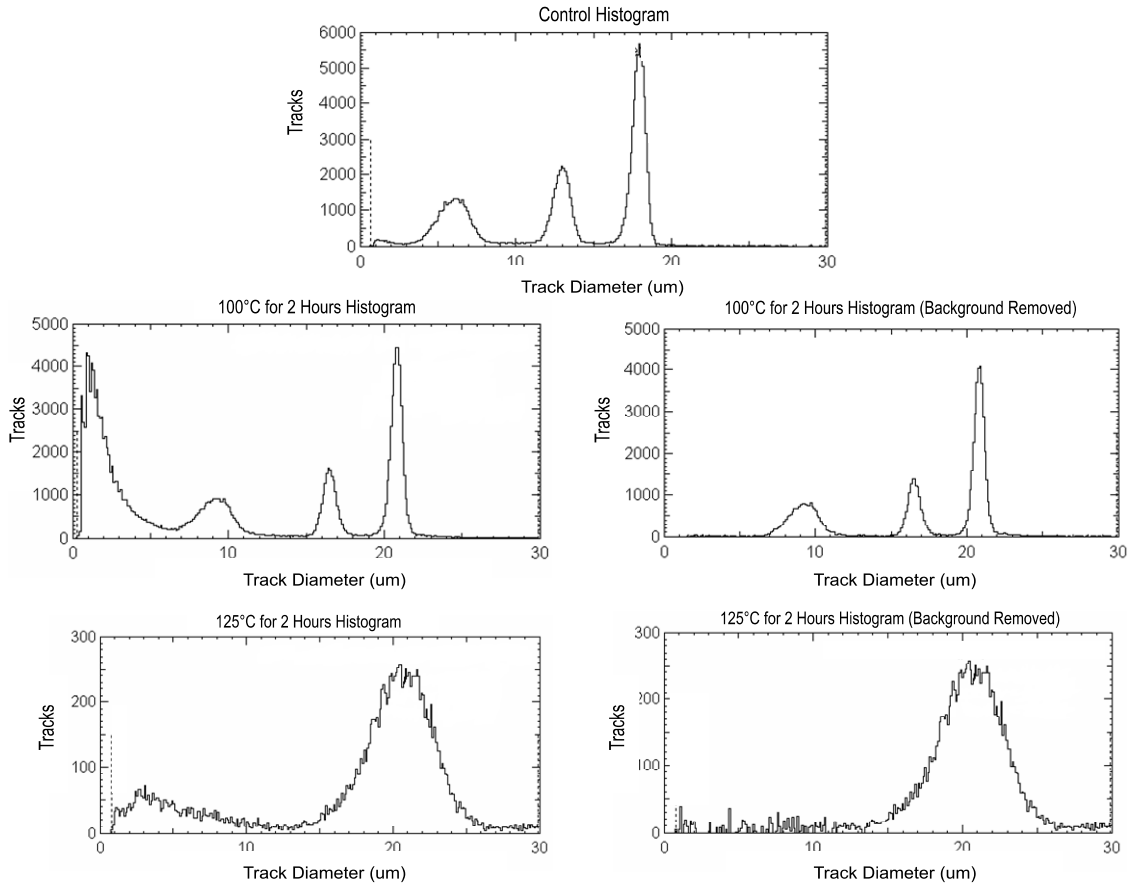


Figure 3: Collected data histograms
The histogram of the control is shown at the top. On the left are histograms including background noise. This noise is seen as a peak at the lower end of the x-axis. Across from these histograms are plots of data from the same CPS scans. Tracks of diameters and depths assumed to be background noise were removed in these histograms.

Diameters at which these peaks occurred were also shifted, correlating to particle energies outside of the range to which samples were exposed. Measured diameters from samples heated at 125°C for two hours prior to exposure identified particles of energies too low to be registered by CR-39 as impacting the detector. Data from samples heated at 150°C for two hours and at 125°C for six hours indicated exposure to particles of only one energy as well, 6.947 and 7.526 MeV respectively. Heating samples for longer periods, including 24 hours at 100°C, resulted in loss of all data. These results are shown in Figure 4.

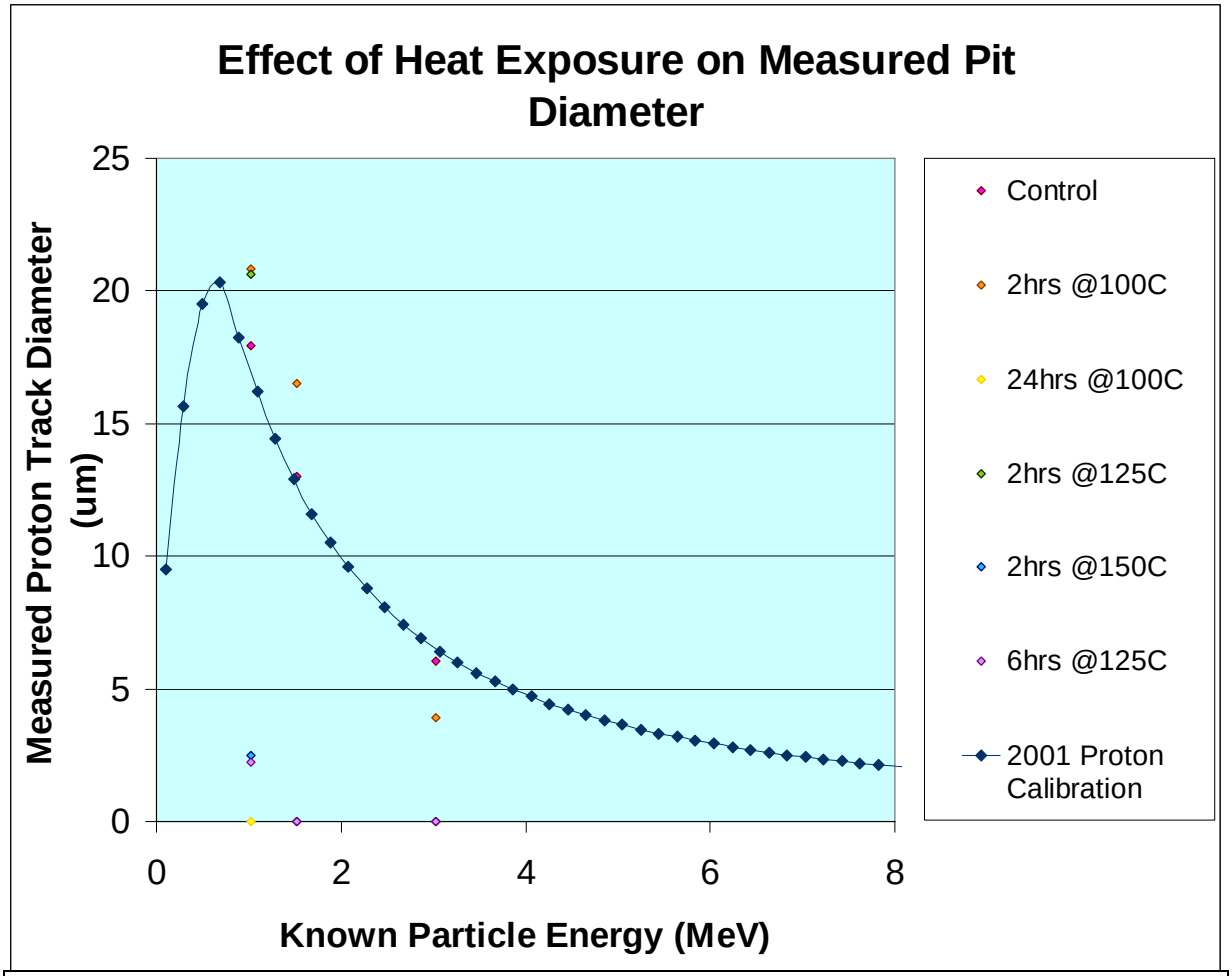


Figure 4: Effect of heat exposure on measured pit diameter
Values on the x-axis represent particle energies as defined by the particle accelerator at SUNY Geneseo. Values on the y-axis represent pit diameters measured using the CPS analysis program developed at MIT. Implied energies can be found by documenting the point at which the "2001 Proton Calibration" curve and a line represented by y=measured pit diameter in μm intersect. The x-value given for this point of intersection is the energy with which particles forming measured pits will be assumed to have impacted the detector surface when data is analyzed.

3. Background Noise

In addition to a change in pit diameter, increased background noise was observed in heated samples (see figure 3). This noise can sometimes be difficult to distinguish from legitimate particle tracks. Little background noise was observed in the control sample, while in heated samples the number of background sites accepted by the CPS system was large in proportion to the number of particle tracks accepted.

It was possible to remove some of this background noise. A CPS scan was taken of an unexposed area of each heated sample. Pits of depths and diameters similar to those accepted in the scan of the unexposed area were then removed from the data set. The results of this process can be seen in Figure 3. Though this method proved successful in removing most of the background noise, some legitimate data may also have been rejected.

4. Testing for Heat Exposure

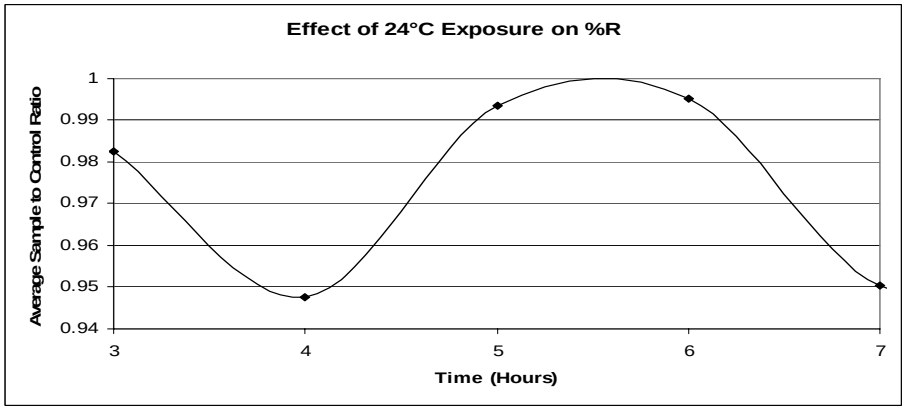
As background noise was observed to increase with heat exposure, it was speculated that surface roughness also increased. All samples were cleaned with ethanol before processing in order to avoid interference from dust particles on the surface of diagnostics. For this part of the experiment, additional detectors were exposed to temperatures ranging from 24°C to 100°C ±1°C and etched in the same manner as the previously described samples. These pieces of CR-39 were not exposed to accelerated particles. Light transmission and reflectance properties of all samples were measured in order to find a simple test to suggest heat exposure. Such a test could be used to identify compromised diagnostics before using these samples on OMEGA or other laser systems, saving time and resources.

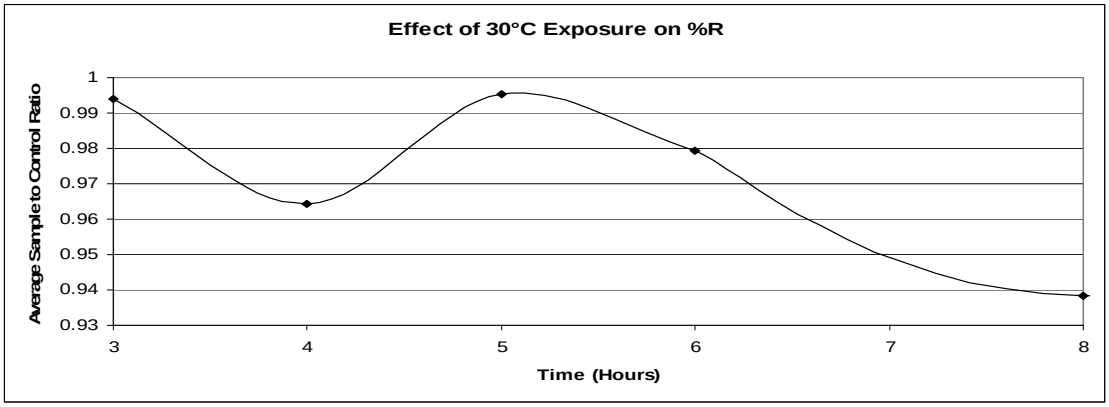
Samples heated at temperatures greater than 100°C could visibly be seen to oscillate between clear and cloudy as a function of time. To quantify this observation, the ability of each

sample to transmit light between wavelengths of 200 and 800 nm was measured by means of a spectrophotometer. Cloudy pieces would absorb more light than clear pieces. Sample transmission measurements were then compared to transmission measurements taken from an etched control. The sample-to-control ratio of percent transmission was seen to consistently oscillate as a function of time exposed. However, this oscillation was of a small magnitude. Consequently, it was necessary to seek another test in which results would demonstrate change much greater than that which could be attributed to measurement error.

The reflection properties of each sample for light of wavelengths of 200 to 800 nm were measured by means of a reflectometer. All samples were again cleaned with ethanol before measurements were taken. Samples were then attached to triangular prisms with a silicone index matching solution. This would limit measured light to only that reflected off the outer surface of each sample, eliminating anomalies caused as light passed through the plastic. By applying index matching solution, light that would normally be reflected upon reaching the plastic-air boundary, coming out of the sample, was made to pass from sample to index matching solution to prism without reflecting and then reflect off the interior prism walls at oblique angles. This light would consequently fail to return to the detector in the reflectometer and would therefore not affect measurements.

The baseline for measurements to be taken from was set with an unheated and unetched detector. The reflectance of each heated sample and of a control was then measured. All heated sample measurements were then compared to control measurements (see Figure 5). Heat was seen to significantly affect the relationship between a control sample's reflectance and that of a heated sample. By etching the frames surrounding detectors during shipment and comparing the reflectance properties of these pieces to the reflectance properties of a control it may be possible





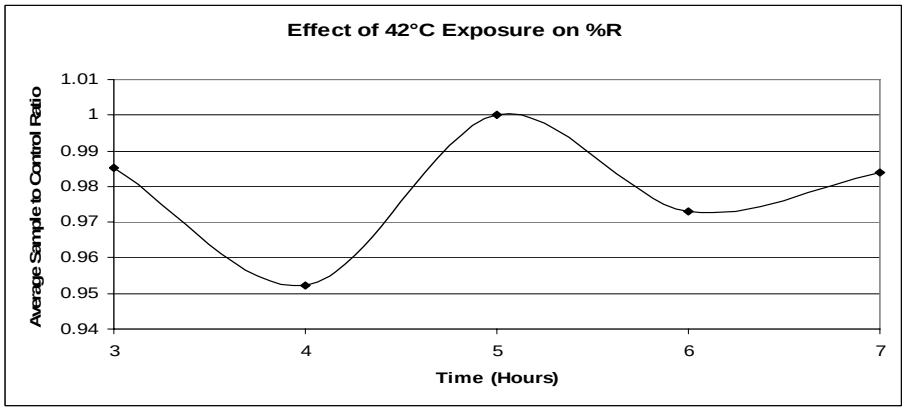


Figure 5: Change in sample-to-control reflectance ratio

to identify compromised pieces prior to their exposure on the laser system. Those pieces with sample-to-control reflectance ratios deviating greatly from one will be rejected before being used on the OMEGA laser system. However, further research is necessary to create more accurate

curves for comparison and a method of testing for heat exposure in those pieces where the sample-to-control reflectance ratio has oscillated back to a value close to one.

5. Conclusions

Exposure of CR-39 to high temperatures clearly affects the accuracy of data collected from these detectors after exposure on the OMEGA laser system. Background noise resulting from heat exposure may inadvertently be accepted as data and thus considered the track of a particle ejected during an attempted fusion reaction. At the same time the tracks of real ejected particles are lost due to changes in properties of the plastic detectors. Heat exposure also causes measured diameters and depths of tracks to deviate from expected values, resulting in inaccurate identification of ejected particles. One simple test for prior exposure to high temperatures is the comparison of the reflectance properties of unused frames to reflectance properties of a control sample. Such frames are made of the same material as detectors and are exposed to the same conditions as the detectors they surround during shipping.

6. Acknowledgements

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7. References

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