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ABSTRACT

The effects of radiation on liquid scintillating fibers composed of borosilicate glass (Pyrex) tubing in combination with a high refractive index liquid scintillator are investigated. The fiber components (tubes, liquids and liquids in tubes), were irradiated in a nuclear reactor with doses exceeding 100 megarads. It was found that the discoloration of the glass tubing does not degrade the optical properties of the fibers - in fact, the attenuation length of fibers using irradiated tubes are longer than those using non irradiated tubes. The attenuation lengths of the liquids included in this study appeared to be shortened by the radiation. However, it was found that no deposits or other surface damage occurred on the surfaces of the tubes containing the liquids so the irradiated fibers could be rejuvenated by simply adding fresh liquid.

KEYWORDS

Liquid scintillator; scintillating fibers; radiation resistance

INTRODUCTION

Scintillating fibers are being strongly considered for use in tracking and calorimetry in both the SSC and LHC experiments. However, the radiation levels are expected to be extremely high in certain areas such as near the beam pipe and in the forward regions (Groom, 1989). Here the doses will be measured in gigarads, whereas existing plastic scintillating fibers can tolerate up to only a few megarads before their performance drops to an unacceptable level. As a possible solution to this problem, the use of fibers employing a liquid scintillator for the core, and a lower index channel or tube for the cladding are being considered.

These "liquid scintillating fibers" are believed to be much more resistant to radiation than the solids for several reasons:

1) Liquid scintillators are inherently less sensitive to certain types of radiation damage than solids and measurements (Zorn *et al*, 1990) confirm this at least for some liquids.

2) Being fluids, liquids can be circulated, allowing *in situ* replacement of damaged material.

3) Early measurements of the effect of radiation damage to the cladding (Datte, 1990) indicate that discoloration has little to no effect.

In addition to concerns for the liquid and cladding separately, another type of possible damage is degradation of the inner surface of the tubing caused by a radiation induced interaction between the liquid and the channel. For example there may be some sort of erosion or coating formed as components of the liquid are broken up by the ionizing particles.

In this study, the effects of radiation are investigated for the specific case where the cladding is Pyrex tubing in combination with a high refractive index liquid. The irradiation was carried out at a nuclear reactor to a dose exceeding 100 megarads. The study addresses all aspects of the problem including the effect on the liquid, the cladding and the combination of the two.

MATERIALS

The fibers used in this study employed standard size Pyrex tubing from Corning. The tube dimensions were 120 cm (48 inches) in length with 2.0 +/- 0.15 mm outer diameter (OD) and 0.5 +/- 0.1 mm wall thickness. This leaves a nominal inner diameter of 1 mm. These tubes were produced by extrusion and resulting surfaces appear smooth down to fractions of a micron under an electron microscope. The diameters, however, were observed to vary significantly. Those with large variations were partially eliminated by requiring a silica optical fiber used for light coupling to fit snugly in both ends.

The tubes were cleaned by rinsing first with distilled water, then reagent grade isopropyl alcohol and finally UV grade acetone. This was found to make a significant difference in the attenuation length compared to not rinsing at all. The cleaning was seen to improve the propagation of the shorter wavelength light most.

The liquid scintillator appearing in all plots presented here was supplied by Bicron (BC-599-13). Three other liquids were tested in parallel including a very similar formula from Bicron and two from National Diagnostics. The second Bicron sample was very similar to the one presented, but had a slightly shorter attenuation length. These liquids have an emission peak near 440 nm and are based on isopropyl biphenyl. The two National Diagnostic liquids had emission peaks near 420 nm and were based on Decalin and Pseudocumene. The attenuation lengths of these liquids were slightly less than those of both Bicron liquids. All four liquids behaved similarly otherwise, and all had refractive indices around 1.57.

IRRADIATION OF SAMPLES

The samples were irradiated at a nuclear reactor at Texas A&M University. The core of the reactor is approximately a cube with 17 inch sides as illustrated in Fig. 1. It sits 5 feet above the bottom of a tank of water, 30 feet in depth, and has a peak operating power of one megawatt. Three of the horizontal sides include graphite shields which reduce the neutron flux in those directions.

The samples were held in a long aluminum tube that could be lowered to the core and placed in position on a side containing the graphite shielding. Perpendicular to the center of the core, the dose rate was measured with an ionization chamber to be about 5 megarads per hour at peak operating power. The rate drops off slightly above and below the centerline, and falls rapidly above and below the upper and lower edges of the core. The samples were irradiated at four heights to make the dose as uniform as possible. Each position was irradiated approximately 20 hours and the samples were left next to the core overnight when the reactor was off, adding a significant additional gamma dose.

The total dose over the whole length of the samples is a minimum of 100 megarads and is estimated to exceed 120 megarads over much of the length. Most of the dose is in the form of gamma's. The neutron flux perpendicular to the core is estimated to have been less than or of the order of 10^{10} cm^{-2} $hour^{-1}$. Low energy (thermal) neutrons are mostly excluded by the shield. This flux is only present when the reactor is on, so for each 20 hour position, the total exposure is approximately 10^{11} cm^{-2} or less.

The Pyrex tubes were held in larger (10 mm OD) Pyrex tubes that were closed at the bottom. For the tube-liquid combinations, the liquids were simply poured into the larger tubes along with the 2 mm OD tubes and covered with Parafilm. The tops were not sealed tightly because preliminary tests showed that gas is released during exposure.

During the irradiation, the Parafilm turned yellow and cracked allowing the liquid to be exposed to the open air. (Components of the Parafilm may have contaminated the liquid as it degraded.) The Pyrex tubes turned a dark brown and the liquids took on a yellowish tint. At such high dose rates, the deposited energy heats the samples, but the temperature was not measured. The reactor operators stated that based on previous

experiments, the temperature could be assumed to be less than the boiling point of water. The Pyrex was slightly activated during the irradiation due mainly to sodium impurities in the glass. The active nuclei had a half life of about twelve hours and was measured to have an activity of less than 0.5 millirads/hour after five days.

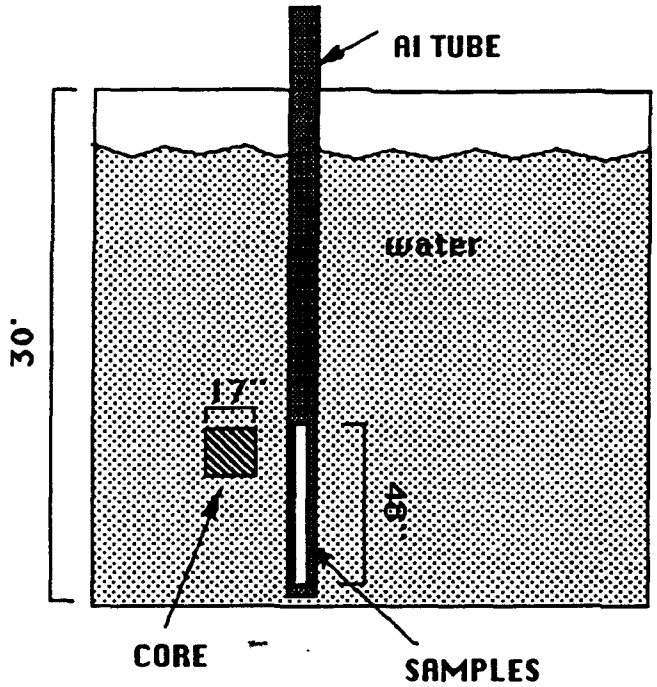

Fig. 1. Layout of sample placement in the nuclear reactor tank.

MEASUREMENTS

The main quantity of interest in this study was the behavior of the attenuation length of the fibers before and after irradiation. The attenuation length was determined by measuring the light output as a function of distance using a test stand as illustrated in Fig. 2a. The liquid fiber is placed in a groove with one end approximately one inch from the face of a photo multiplier tube (EMI 9839b). The fiber is inserted in a small hole in a shield in front of the photo tube to allow light only from the core to be measured. Optical filters can be placed in a slot between the fiber and the photo tube. A 50 millicurie Sr^{90} source is driven by a stepper motor along the sample in two inch steps and the photo tube current is read out twice at each point. The current is measured by a Keithley 485 picoammeter which is read out through an IEEE-488 port. A PC controls the stepper motor and reads the picoammeter. The dark current is 1-3 nA and the signal varies from about 1 μA to as low as 50 nA when using a 515 nm optical filter. The groove containing the sample is aligned relative to the motion of the source to within 0.005 inches (1/8 mm).

The samples are filled by drawing the liquid into the glass tubes using a syringe which is coupled to the glass with a flexible (Tygon) tube. The liquid is allowed to touch only the glass to assure minimal contamination. A silica optical fiber (3-M Power Core), with a 1 mm OD and 2 inches in length, is inserted into one end and glued with 5-minute epoxy as illustrated in Fig. 2b. The optical fiber has a 600 micron silica core with a silicate cladding and a Tefzel (3-M) jacket. These materials were tested for chemical compatibility with the liquids and appear to be non-destructive. The other end is also sealed with a drop of 5-minute epoxy. An air gap prevents the epoxy from touching the liquid on this end while contact with the liquid on the fiber end is minimized by inserting the fiber approximately 1/2 inch into the tube and wicking away excess liquid before gluing. This method allows the coupling to be made without an air bubble between the liquid and the silica fiber. If a small air bubble is trapped, it can be removed by allowing it to rise to the end with the air gap.

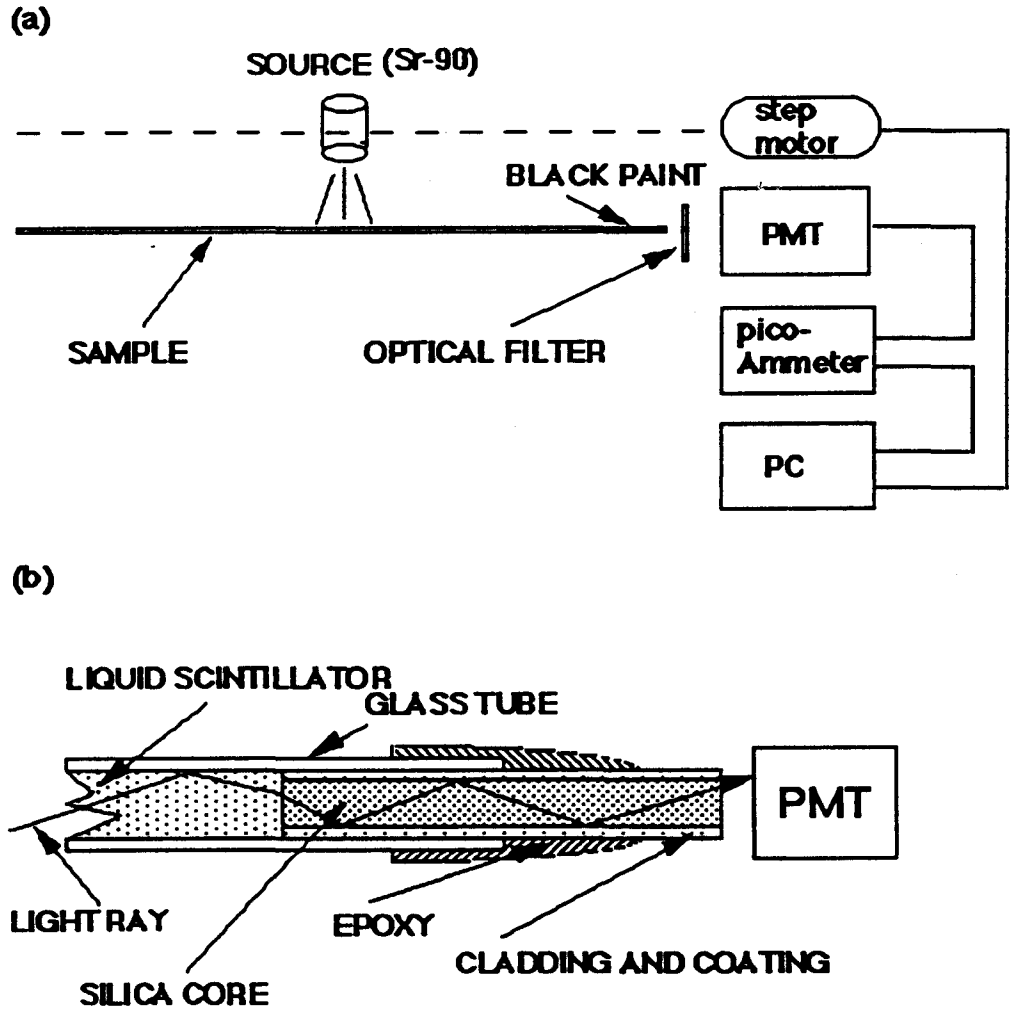

Fig. 2. a) Test stand for measuring attenuation lengths: b) Light coupling of liquid fiber to photo tube using a silica core optical fiber.

After the glue is dry, the end of the Pyrex tube containing the silica fiber is painted with a graphite paint (Aquadac). The paint covers approximately two inches including the epoxy and a small part of the silica fiber. This paint is necessary to eliminate the light trapped in the air-glass interface.

RESULTS

An assortment of fibers were constructed including combinations of non irradiated glass with fresh liquid, non irradiated glass with irradiated liquid and irradiated glass with fresh liquid. Several samples of each type were made to study variations. An example of light versus distance curves for several samples of BC-599-13 in non irradiated tubes is shown in Fig. 3. The photo tube sits approximately 5 inches to the left of the first data point on these plots. The variation in average light output between samples is due mainly to variations in light coupling. The silica fibers were "cut" by simply scratching and breaking, leaving variations in the surfaces. Also, the distance from the end of the fiber to the photo tube was not precisely fixed. There is also a small effect from variations in the average tube diameter. The shapes (slopes) of the curves are seen to be quite similar, however. The attenuation lengths determined by a fit to an exponential function including all points are, from the top curve to the bottom, 1.58, 1.58, 1.57, 1.57 and 1.52 meters respectively.

If the fit is restricted to the points further from the tube (distances greater than 17 inches, for example), the attenuation lengths are significantly longer. However, the small bumps and wiggles which are due to

fluctuations in tube diameter prevent an accurate determination of the slope once it becomes this small. In this case, the attenuation lengths are, from top to bottom, 1.85, 1.69, 1.83, 1.88 and 1.97 meters respectively.

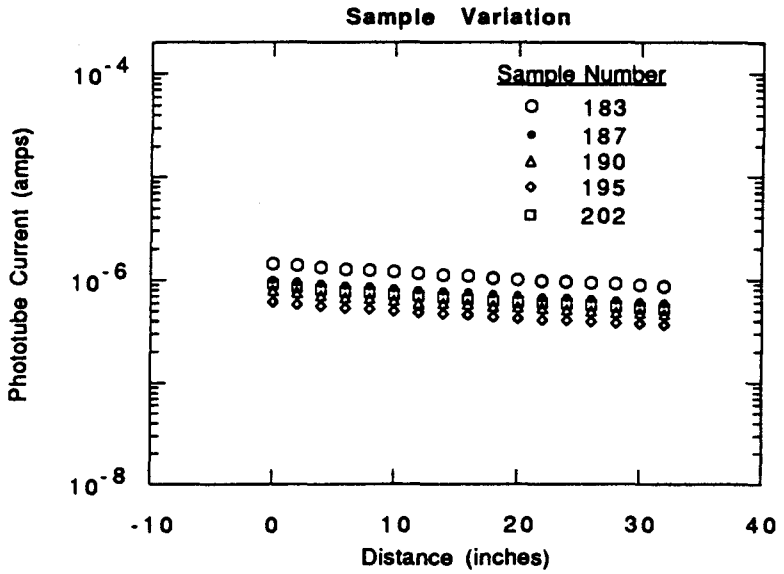

Fig. 3. Light versus distance curves for five fibers made of non irradiated glass and BC-599-13 scintillator.

The effect of the radiation damage to the glass tubes (cladding) is illustrated in Fig. 4. In this figure, the light versus distance curves are shown for two different fibers as a function of optical filter. The open symbols represent a fiber with non irradiated glass and fresh liquid. The top most curve has been normalized so that the data point at the distance "0" is scaled to a light output of "1". The curves for the three filters are then scaled by this same factor. The measurements were done by first measuring the fiber with no filter and then by adding filters without disturbing the fiber, so the relative light outputs are a true indication of the reduction of light as a function of filter. (The filters are "high pass filters" in which only light with a longer wavelength than the label is passed.)

The solid symbols define the curves for a fiber composed of an irradiated glass tube with fresh liquid. Again, all curves were scaled as was done for the non irradiated fiber. Note that the relative light outputs of the two fibers are almost identical and that the slopes of the curves for the tube made of irradiated glass are slightly less than those of the non irradiated tube.

The attenuation lengths as determined by an exponential fit to the entire curves are, from top to bottom, 1.6, 2.0, 2.7, and 2.8 meters respectively for the tubes with non irradiated glass, and are 1.8, 2.9, 3.7 and 4.2 meters respectively for the tubes made of irradiated glass. However, as discussed, these numbers are sensitive to the bumps from diameter variations, and cannot be considered entirely accurate.

In order to study the radiation damage to the liquid scintillator, fibers were constructed using irradiated liquid in combination with non irradiated glass. Figure 5 shows a comparison between one of these and one with fresh liquid. The attenuation length of the damaged liquid fiber is approximately 20 cm, in contrast with 1.6 meters for the non irradiated fiber. In this plot the liquid is BC-599-13, but all the liquids had similar drops. The light output of the damaged liquids were measured in a "liquid scintillation counter". Compared to non irradiated liquids, the drop was only about 20%.

As stated, the glass tubes were irradiated in both an air atmosphere and in the various liquids. The irradiated glass shown in Fig. 4 was irradiated in air. The ones irradiated in liquid were rinsed with water and isopropyl alcohol to flush the damaged liquid and filled with fresh liquid. The light versus distance curves were found to be identical to the ones for the tubes irradiated in air within tube variations. This suggests that

no surface damage (coating or other) is caused by interactions between the liquid and glass under the conditions in which the study was done. Samples of the glass were also inspected under an electron microscope and no surface changes were observed. Also, visually, there appeared to be no difference between those irradiated in liquid and those irradiated in air.

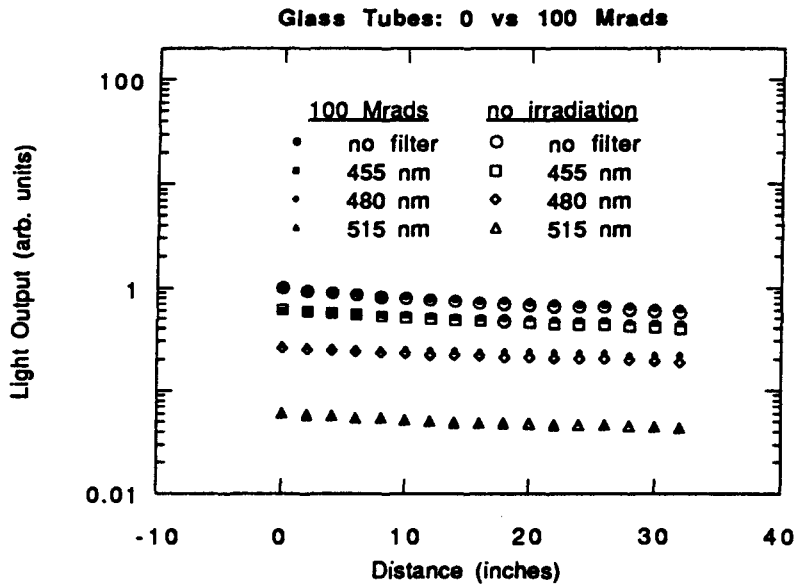

Fig. 4. Light versus distance curves comparing a fiber made of non irradiated glass with one made of irradiated glass. Both had fresh liquid. Each fiber is measured with a series of optical filters (none, 455 nm, 480 nm and 515 nm) in front of the photo tube.

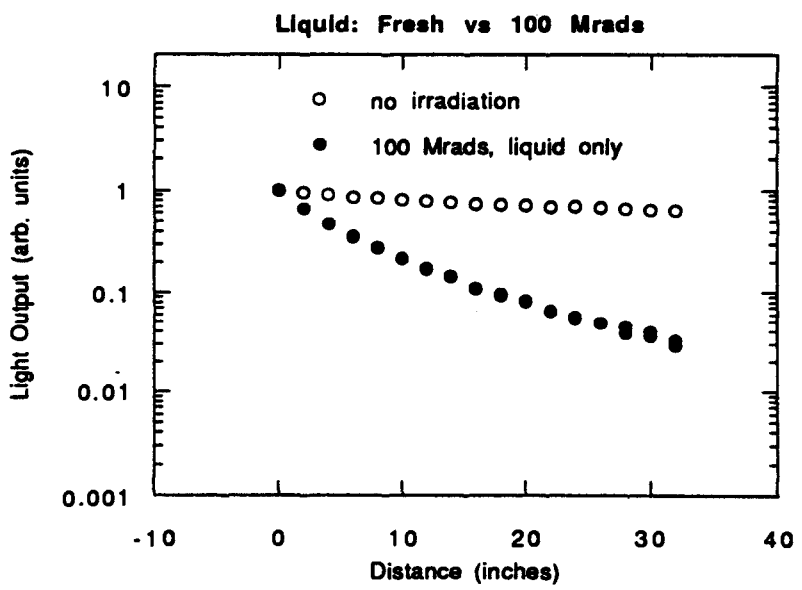

Fig. 5. Light versus distance curves comparing a fiber constructed out of irradiated liquid with one using fresh liquid. Both had non irradiated glass cladding.

SUMMARY

The main findings of this study can be summarized as follows:

1) For liquid scintillating fibers using Pyrex tubing for the cladding, there appears to be no degradation of the attenuation length for doses over 100 megarads. Instead, the attenuation length of the fibers appears to be longer after irradiation of the tubing.

2) Under the conditions of the test (air atmosphere, high dose rate, Parafilm cover), the liquids were degraded with attenuation lengths decreasing by a large factor, but light output dropping by only 20%.

3) No coatings or other surface degradation were observed in tubes irradiated in the liquids. This was checked visually, by electron microscope and by measurement of the attenuation lengths.

The apparent increase in attenuation length after irradiation of the tubes is possibly due to a further cleaning effect. As stated earlier, it was found that the attenuation length increased significantly if the surface was cleaned chemically. It was also found that the cleaning had the greatest effect on the shorter wavelengths as expected for Raleigh scattering. Such scattering due to surface imperfections and even density or dielectric fluctuations is a major cause of attenuation in communications fibers. To test whether or not the effect could be explained by the radiation having an annealing effect on the glass that could reduce dielectric fluctuations, a sample of tubes were annealed at 520 degrees Celsius for 16 hours and irradiated along with the non-annealed tubes. No difference was observed between the two, suggesting that some other effect is responsible.

The damage to the liquids in this test is not surprising. The liquids were exposed to air (and ozone) during the irradiation and for several days afterwards. They were also at an elevated temperature during exposure to the radiation. The yellow appearance is suggestive of the formation of oxygen compounds according to both Bicron and National Diagnostics. The tests will be repeated in a more controlled atmosphere to check this hypothesis.

Finally, the fact that no surface interactions between the liquids and the glass were observed is important since such an effect would have greatly complicated the issue of whether or not liquid scintillating fibers are sufficiently "rad-hard" to be usable in highest radiation environments at the SSC and LHC. The lack of deposits or other damage means that the liquids can simply be replaced by circulation as degradation occurs.

These studies are continuing and expanding to include more liquids, other types of tubing (longer and more uniform) and higher doses. The current best liquid with a 480 nm filter has an attenuation length in Pyrex tubing approaching 3 meters, and is even longer for the irradiated tubes. This is well within the range needed for many applications, such as in the forward calorimeters at the SSC and LHC.

ACKNOWLEDGMENT

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