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NASA Grant NGR 07-010-002

to

Fairfield University, Fairfield, Conn.

"Investigation of Energy and Deposition Processes in the Upper Atmosphere and the Interaction Between the Mesosphere and the Thermosphere"

Principal Investigator - Dr. James H. McElaney

This grant was awarded on March 1, 1968 for a period of one year. The terminal date was later extended to June 1, 1969.

The original purpose of the grant was to examine the airglow and nightglow through a series of ground based spectrophotometric measurements which were to be correlated with data telemetered from rockets launched into the aurora. The experiments were to be performed at Fort Churchill, Resolute Bay, and Wallops Island. The experimenter was to provide measurements at the time of, and in support of, these rocket launches. The measurements were to cover the broad spectral region of 3000 Å to 8500 Å. On April 15, 1968, after the spectrophotometer had been previously calibrated, a trip was made to Fort Churchill to perform the ground based measurements in support of the rocket launch. Unfortunately, seconds after launch the booster rocket exploded, and the rocket instrument package was lost. Consequently it was impossible to correlate ground based measurements with the rocket data.

It was then decided to undertake work related to the use of optical materials used in experiments aboard rockets or satellites.

A problem of particular interest was the effect of proton irradiation on optical materials. Work was begun on this project and the transmittances of 19 optical crystals was measured between 1050 Å and 3000 Å. These measurements were carried out on a McPherson 1 meter vacuum spectrograph. The transmittances were measured before and after irradiation by 5 MeV protons with a flux of 10^{12} protons/cm^2. This radiation corresponds to the energy and flux expected for exposed optical materials in a Nimbus type spacecraft in a circular orbit at an altitude of 1200 km. This work has been completed and was initially submitted for an in-house publication. It has been revised and is being submitted for publication to Applied Optics. A copy of this article is attached.

Permission was requested from NASA Headquarters for a time extension of one month, and for the expenditure of grant funds for purchase of equipment to be used in the ground support program, for laboratory equipment needed for the calibration of the instrument, and other equipment subject to the provisions of the grant instrument. This permission was granted on February 19, 1969. Accounting records have been kept according to accepted practices and are available on request.

Work is now being done to calibrate a 1 meter Ebert-Fastie spectrophotometer, F/4, with a 95 cm focal length telescope. This instrument will scan the spectrum continuously with a scanning speed of approximately 15 seconds. This instrument will be used for spectrophotometric measurements of the airglow and nightglow to be made later this year. This work will be done under the terms of the funding granted for the coming year, Grant NGR 07-010-002 (Supplement).

THE EFFECTS OF PROTON IRRADIATION ON THE ULTRAVIOLET TRANSMITTANCES OF OPTICAL MATERIALS BETWEEN 3000 Å AND 1050 Å

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I. INTRODUCTION

A great deal of work has appeared in recent years in the literature of solid state physics on the affects of high energy radiation on the optical properties of crystals and glasses. The purpose of much of the work performed was to irradiate the crystals or glasses and produce crystal defects and color centers. Most of this work, therefore, required that the irradiation dose be much greater than would be experienced by optical materials in an earth orbit during the course of a year. In addition, most of the work has been limited to the nonvacuum region above 2200 Å.

The purpose of this present work is to investigate the effects on the transmittance of optical materials commonly used in the ultraviolet when subjected to irradiation with high energy protons. The radiation dose is what might be encountered in space in a Nimbus satellite orbit in the course of a year. This study is a continuation of the work of Heath and Sacher on the effects of high energy electrons on the transmittance of optical materials.[1] It is also a continuation of, and supercedes, the work of Sacher on the effects of proton irradiation on optical materials.[2] This study has been limited to materials which are transparent in the ultraviolet region of the spectrum, from 1050 Å – 3000 Å.

The materials studied in the work are listed in Table 1. All of these materials are frequently used in the far ultraviolet and vacuum ultraviolet regions of the spectrum.

II. PROTON ENERGY DISTRIBUTION IN SPACE

According to Vette[3], the trapped inner zone proton radiation belt is relatively stable in time, however minor changes have been observed due to changing atmospheric conditions. An analysis was performed to determine what the total trapped proton radiation environment would be for a Nimbus type spacecraft for the year 1968 in a circular polar orbit at an altitude of 1100 km. It seems reasonable to simulate, at least in part, the radiation of the space environment by irradiating the crystals under investigation with 5 MeV protons at a flux of 10^{12} protons/cm^2.

TABLE I. OPTICAL MATERIAL CHARACTERISTICS

Crystal	a (Å)	b (mm)	c (Å)	d
LiF	1050	3.25	1300-3000	amber
MgF_2	1130	3.05	–	amber
CaF_2	1210	1.00	–	clear
NaF	1250	3.20	1260-3000	clear
LaF_3	1260	1.45	e	amber
SrF_2	1280	3.20	–	clear
BaF_2	1340	3.35	–	clear
Al_2O_3	1435	3.20	–	amber
Corning 7940	1580	3.30	–	clear
Dynasil #1000	1590	3.30	–	clear
Suprasil	1590	3.30	–	clear
ADP	1590	3.15	–	clear
NaCl	1750	3.20	1850-2250	amber
KCl	1800	3.40	–	violet
CdF_2	1990	2.00	–	amber
KBr	2010	3.20	–	blue
Calcite	2040	3.00	e	amber
Corning 9-54	2200	3.25	All	amber
Corning 7-54	2270	3.05	All	clear

a. Ultraviolet transmission limit.
b. Thickness.
c. Radiation-induced absorption features.
d. Color after irradiation.
e. Phosphoresced.

III. EXPERIMENTAL PROCEDURE

The transmittances of the crystals were measured at the exit slit of a 1-m McPherson Model 225 monochromator using a Hinteregger-type windowless light source. A sample holder for the crystals was so constructed that the crystal could be inserted or removed from the exit beam without breaking the vacuum.

The source for protons was a tandem Van de Graaff accelerator located at the Naval Research Laboratory, Washington, D. C. The crystals were mounted on a metal plate which was placed orthogonal to the proton beam. The crystals listed in Table 1 were then irradiated with 10^{12} protons/cm^2 at 5 MeV. The irradiation time was approximately 45 seconds.

The measurements of the transmittances were all made within a week and a half after irradiation. Each crystal was examined for phosphorescence within several hours of the irradiation.

IV. TRANSMISSION CHANGES DUE TO PROTON IRRADIATION

A. Alkali Halides

a. Lithium Fluoride

LiF is the most commonly used window material in the vacuum ultraviolet region. It has the shortest wavelength transmission (1040 Å) of any crystal. The transmission properties of LiF deteriorate when the crystal is exposed to moist air or when the crystal is exposed to vacuum ultraviolet radiation.[4] Warming the crystals to about 500°C usually restores the original transmittances. Cleaning the crystals with ethyl alcohol in an ultrasonic cleaner can also restore the transmission properties.[5] The transmittances may also decrease when argon or hydrogen discharges are used due to the formation of color centers. However, these color centers do not appear when xenon or krypton discharges are used.[6] The original transmittance can be restored by annealing at 500°C. Heath and Sacher found that an electron irradiated sample of LiF became completely opaque, except for a slight transparency between 1200 Å and 1650 Å.[1]

The sample used was an optically polished, high purity crystal made by Optovac, Inc. The transmittance of the crystal before and after irradiation is given in Figure 1. The transmittance suffered a decrease under proton irradiation from 1300 Å to 3000 Å. In addition, there is a very pronounced and symmetrical absorption band to be noted around 2500 Å. It is apparent that LiF is not well suited for space optics if it is exposed to either electron or proton irradiation.

b. Sodium Fluoride

The NaF crystal which was examined showed a reduction in transmittance between 1260 Å and 3000 Å. The absorption varied from 3 percent at 1700 Å to 15 percent at 3000 Å, as is evident from Figure 2. The absorption seemed to be increasing toward longer wavelengths. There was no change in the color of the crystal after proton irradiation.

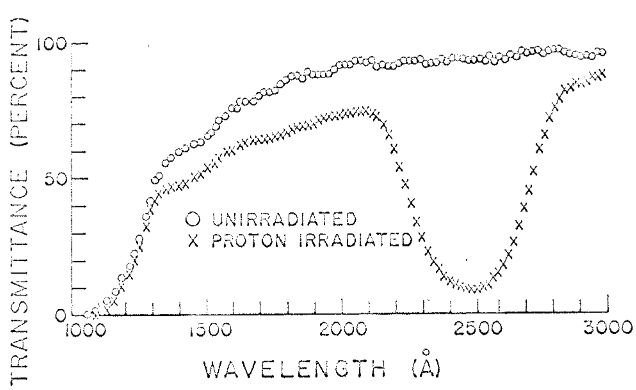

Figure 1—Transmittance of LiF before and after irradiation by a total flux of 10^{12} protons/cm^2 at 5 MeV.

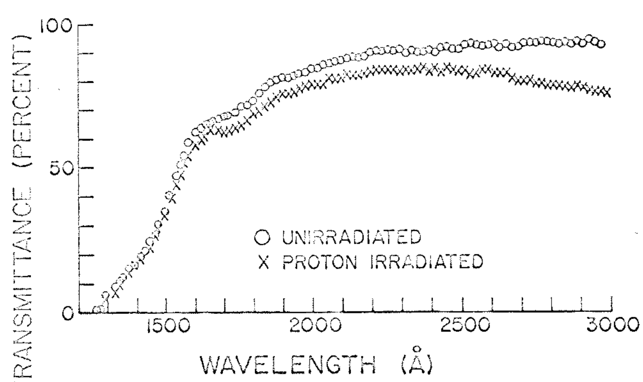

Figure 2—Transmittance of NaF before and after irradiation by a total flux of 10^{12} protons/cm^2 at 5 MeV.

c. Sodium Chloride

The NaCl crystal changes to an amber color after the proton irradiation. It also showed, Figure 3, a decrease in transmission from 1850 Å to 2250 Å.

d. Potassium Chloride

The transmittance of KCl is given in Figure 4. While there is no reduction in transmission after irradiation, the crystal became quite violet after the irradiation.

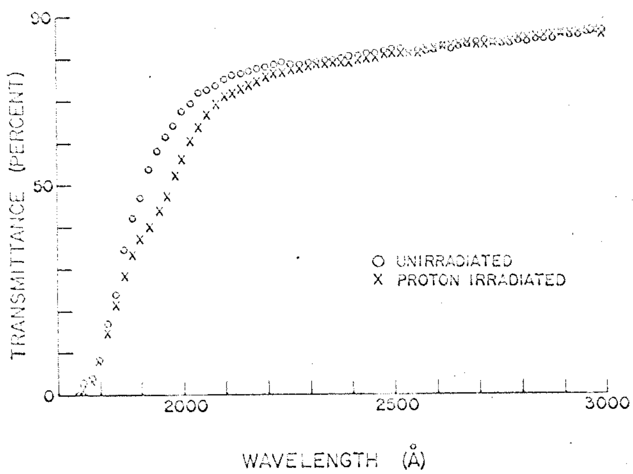

Figure 3—Transmittance of NaCl before and after irradiation by a total flux of 10^{12} protons/cm^2 at 5 MeV.

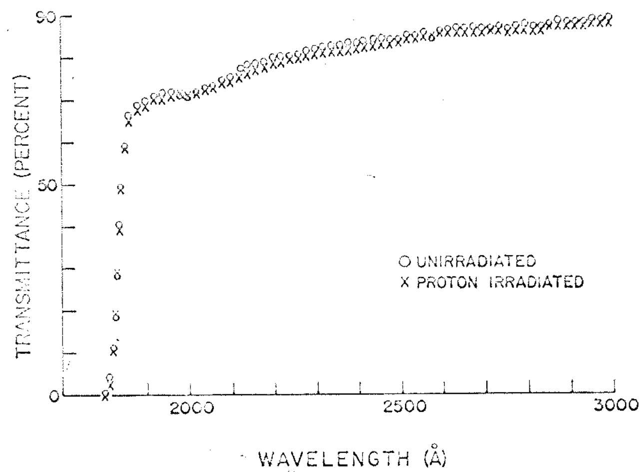

Figure 4—Transmittance of KCl before and after irradiation by a total flux of 10^{12} protons/cm^2 at 5 MeV.

e. Potassium Bromide

Under proton irradiation the KBr crystal noticeably changed its color to blue. No change was noted in the transmittance of the crystal, however, when it was measured two days later. The transmittances for KBr appear in Figure 5.

B. Alkaline Earth Halides

a. Magnesium Fluoride

The sample used was a high purity crystal grown and polished by the Harshaw Chemical Company. It has been found in the past few years that MgF_2 crystals are available which can transmit below 1300 Å. The particular crystal used in this work began to transmit at approximately 1130 Å, as is indicated in Figure 6. The crystal showed no change in transmittance after irradiation.

MgF_2 has the added advantage of being considerably less soluble in water than the LiF (0.013 vs. 0.27 g/100 g of H_2O). It has about the least solubility of any of the fluorides except CaF_2. This is an important feature, since moisture in the air reacts with many of the fluorides, reducing their transmission. Canfield et al. have reported results of extended tests on MgF_2 coatings. They observed no aging effects nor any loss in reflectance after irradiation with 1 MeV electrons and 5 MeV protons. Under electron irradiation two absorption bands developed, a strong absorption band at 2600 Å as well as the absorption band at 1300 Å noted in previous work.[1] Our own investigation showed no appreciable change in the transmittance after irradiation by 5 MeV protons.

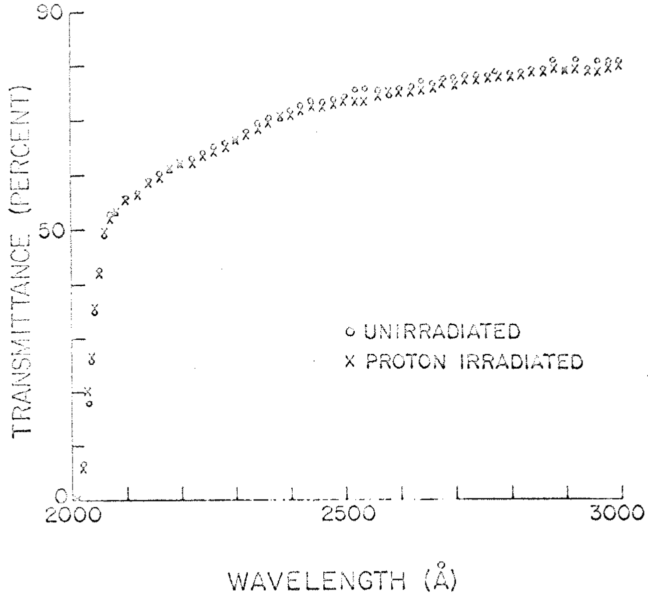

Figure 5—Transmittance of KBr before and after irradiation by a total flux of 10^{12} protons/cm² at 5 MeV.

b. Calcium Fluoride

The crystal sample used was manufactured by the Harshaw Chemical Company. There was no color change noticeable in the crystal after irradiation. CaF_2 does not appear to be sensitive to proton irradiation. In a previous study of electron irradiation of optical materials, two distinct absorption features appear, one at 1900 Å and the second at 2250 Å.[1] These absorption bands do not appear in CaF_2 as a result of proton irradiation. There may be a slight indication of an absorption band at 1950 Å which is present in CaF_2 both before and after proton irradiation. However, the evidence for this may lie within the limits of experimental accuracy.

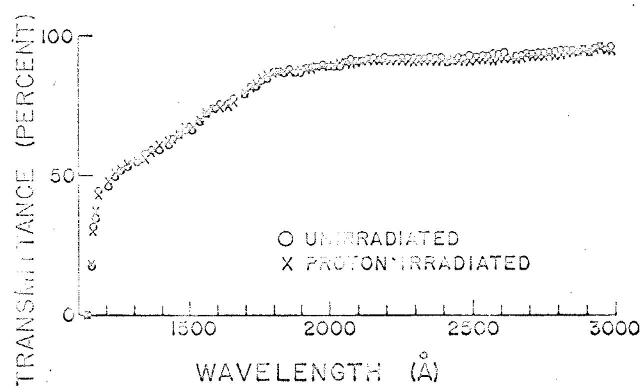

Figure 6—Transmittance of MgF_2 before and after irradiation by a total flux of 10^{12} protons/cm² at 5 MeV.

c. Strontium Fluoride

The transmission curve for the SrF_2 crystal is given in Figure 7. There is no reduction of the transmission after irradiation, nor was there any coloration of the crystal after irradiation.

d. Barium Fluoride

The sample investigated was an optically polished crystal, 3.35 mm. thick, obtained from Optovac, Inc. The transmittances remained unchanged after proton irradiation. Interestingly

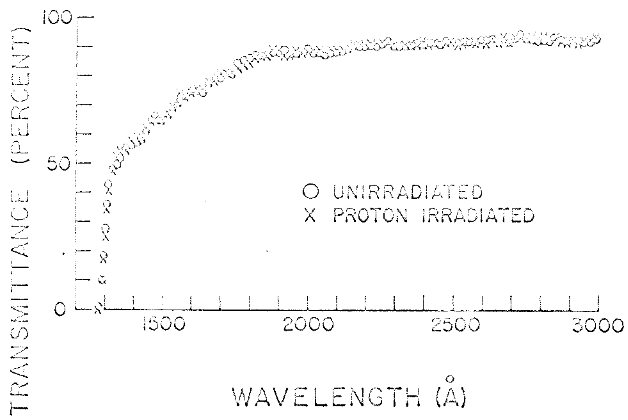

Figure 7—Transmittance of SrF_2 before and after irradiation by a total flux of 10^{12} protons/cm^2 to 5 MeV.

enough, the crystal had an absorption band centered at 1900 Å, before irradiation. Even in this absorption band, no change was noted after irradiation. There also was no color change observed in the crystal after irradiation.

If care is exercised in selecting an optically pure sample, BaF_2 appears to be a good material for space application. BaF_2 may be useful not only as a filter material, but as a low index of refraction element of an achromat for the vacuum ultraviolet.

Messner and Smakula worked on the absorption of BaF_2 colored by 3 MeV electrons at 20°C.[8] Their work showed a major absorption band at 2000 Å and a minor one at 2500 Å. These absorption bands were not noted by Heath and Sacher[1] in their study of optical materials irradiated by electrons, nor are they apparent in BaF_2 when irradiated by protons.

e. Lanthanum Fluoride

The LaF_3 crystal turned an amber color after proton irradiation. The crystal fluoresced slightly when it was examined a few hours after proton irradiation. The fluorescence occurred from 4750 Å - 4950 Å and from 5950 Å - 6150 Å. As is clear from Figure 8, there is no decrease in transmission after proton irradiation. The irregularities occurring in the transmission curve also remained constant after irradiation.

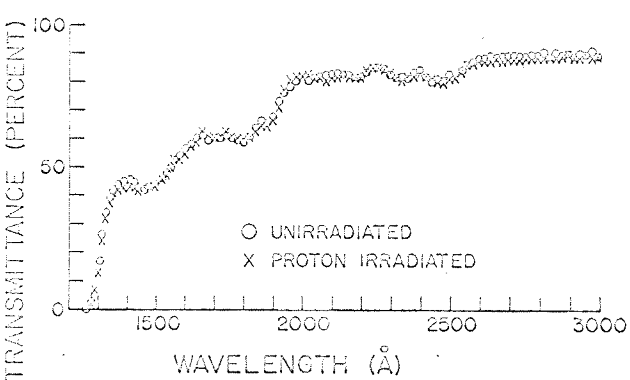

Figure 8—Transmittance of LaF_3 before and after irradiation by a total flux of 10^{12} protons/cm^2 at 5 MeV.

C. Sapphire

This crystal appears to be resistant to proton irradiation, although a change in color was observed immediately after irradiation. Previous work observed only small decreases in transmittance after electron irradiation at about 2600 Å - 2950 Å.[1] The decrease in transmittances at these points after electron irradiation may correspond to induced absorptions by reactor irradiation at 2554 Å and 3000 Å as reported by Levy.[9]

Strangely enough, the well known absorption band at 6.06 eV (2040 Å) is not observed from these measurements. It appears that synthetic sapphire is highly resistant to high energy electron and proton irradiation such as is encountered in the lower regions of the radiation belts. Therefore, Al_2O_3 is useful for shielding optical materials which are sensitive to radiation damage provided that the wavelength limit of transmission of sapphire can be tolerated.

D. Fused Silica

Three samples of fused silica were investigated. The Corning 7940 produced by the Corning Glass Company, and Dynasil #1000 and Suprasil which are produced by Engelhard Industries. These crystals are 3.30 mm. thick, optically polished, and high purity samples. All the crystals have rather sharp ultraviolet cutoffs lying between 1500 Å and 1590 Å. The transmittance curve for Suprasil is indicated in Figure 9. Transmittances for Corning 7940 and Dynasil #1000 are basically those of Heath and Sacher.[1] Towards the upper wavelength regions these crystals seem to have a better than 90 percent transmittance before and after proton irradiation. There were no color changes produced in these crystals as a result of the proton irradiation.

Fused silica, in addition to its desirable optical properties, has the advantage of being able to withstand high temperatures and thermal shock. In addition to natural crystalline quartz, a number of types of fused quartz, fused silica, or quartz glass are widely used for envelopes of light sources and many other laboratory purposes.

From the transmittance measurements made, it is apparent that these samples of fused silica are highly resistant to proton irradiation and appear well suited for space application.

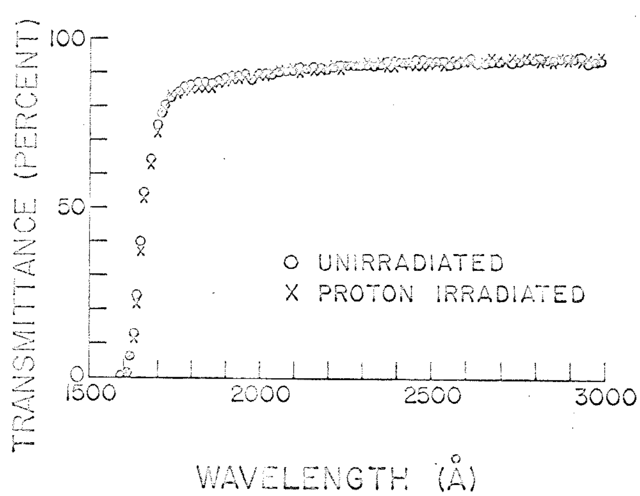

Figure 9—Transmittance of Suprasil before and after irradiation by a total flux of 10^{12} protons/cm² to 5 MeV.

E. ADP

This ammonium-di-hydrogen phosphate crystal, 3.15 mm. thick, was grown and polished by the Harshaw Chemical Company. The crystal has a sharp ultraviolet cutoff at about 1795 Å, and shows no change after being irradiated by protons. There are a number of problems which might be encountered in using this crystal since it is fairly hygroscopic and it is also sensitive to thermal shock. However, there was no noticeable color change in the crystal associated with the irradiation. It may be concluded that ADP is useful for optical use in space below 3000 Å.

F. Cadmium Fluoride

This synthetic crystal from the Harshaw Chemical Company is a sample 2 mm. thick. After proton irradiation the crystal showed a distinct amber coloration. However, as is apparent from Figure 10 the crystal is not sensitive to proton irradiation.

G. Calcite

From Figure 11 it can be seen that this calcite crystal of high optical quality is quite resistant to proton irradiation. The crystal did undergo a noticeable color change, however, after irradiation. In addition, the crystal phosphoresced strongly between 5550 Å and 7000 Å as is indicated by Figure 12.

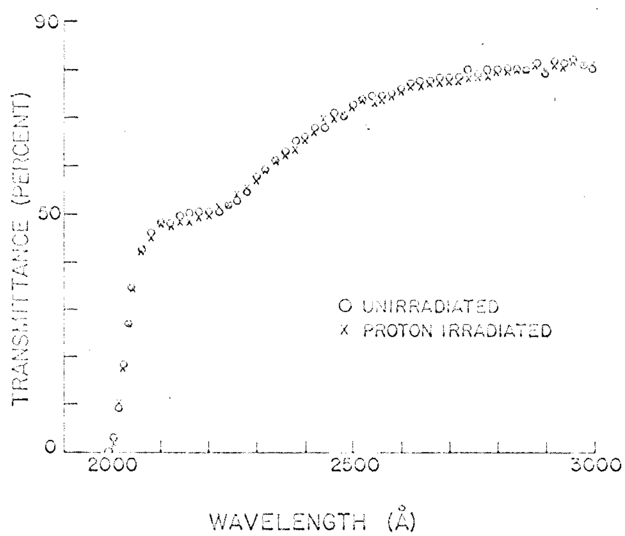

Figure 10—Transmittance of CdF_2 before and after irradiation by a total flux of 10^{12} protons/cm^2 at 5 MeV.

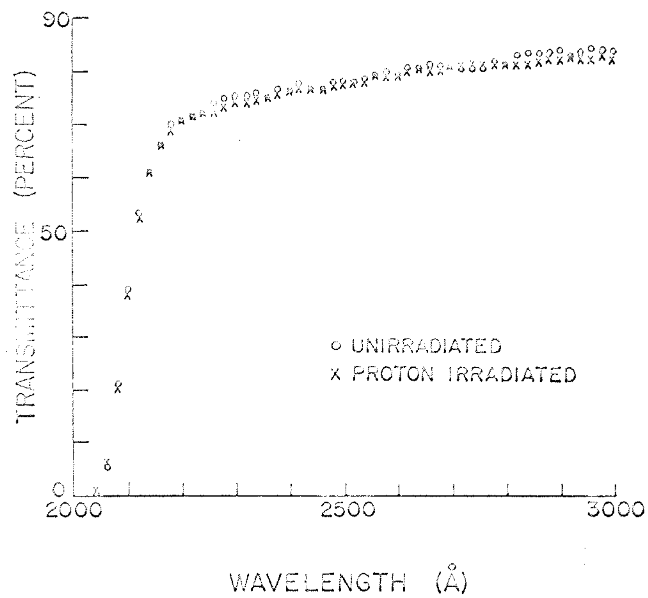

Figure 11—Transmittance of calcite before and after irradiation by a total flux of 10^{12} protons/cm^2 at 5 MeV.

H. Corning Glasses

Two samples of Corning glasses were selected for a study of their transmittances before and after proton irradiation. They are the familiar Corning 9-54 (Vycor 7910) and Corning 7-54. Their transmittances before and after proton irradiation are illustrated in Figures 13 and 14 respectively.

These are high-silica content glasses. They are made by leaching the fluxes from alkali borosilicate glass and firing the residual high-silica porous structure at about 1200°C to consolidate it.[10] The treatment shrinks the porous structure to about 50 percent of its original volume. This process makes the exceptionally high temperatures which are required for quartz unnecessary and makes possible fabrication of a large number of shapes which could not be made from such a high melting glass in any other practical way.

As appears from Figure 13, there is a decrease in the transmission of the Corning 9-54 throughout the wavelength region which was examined. This glass turned an amber color after irradiation.

The Corning 7-54 is the familiar black ultraviolet transmitting glass whose transmittance reduced rapidly in an irradiated environment, as is evident from an examination of Figure 14. Since this Corning glass (9863) also decreases upon exposure to intense ultraviolet radiation, it is evident that it too is a poor material for use in space.

V. CONCLUSION

The purpose of this investigation was to determine the effect of a high energy proton

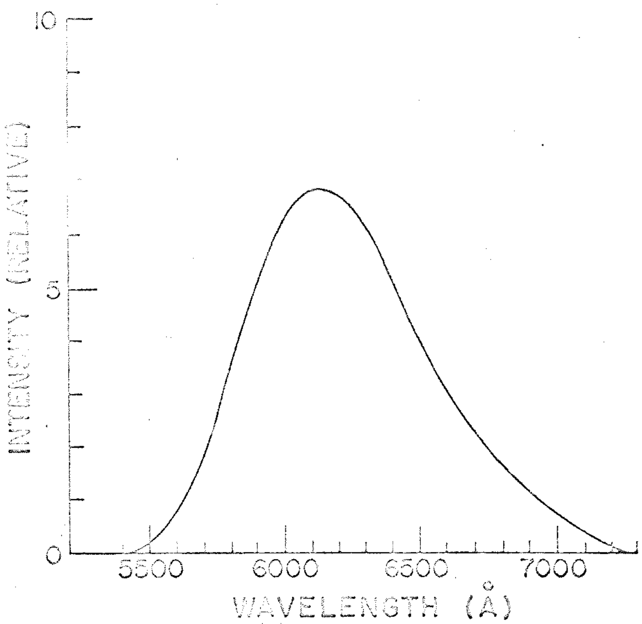

Figure 12—Phosphorescence of calcite after irradiation by a total flux of 10^{12} protons/cm² at 5 MeV.

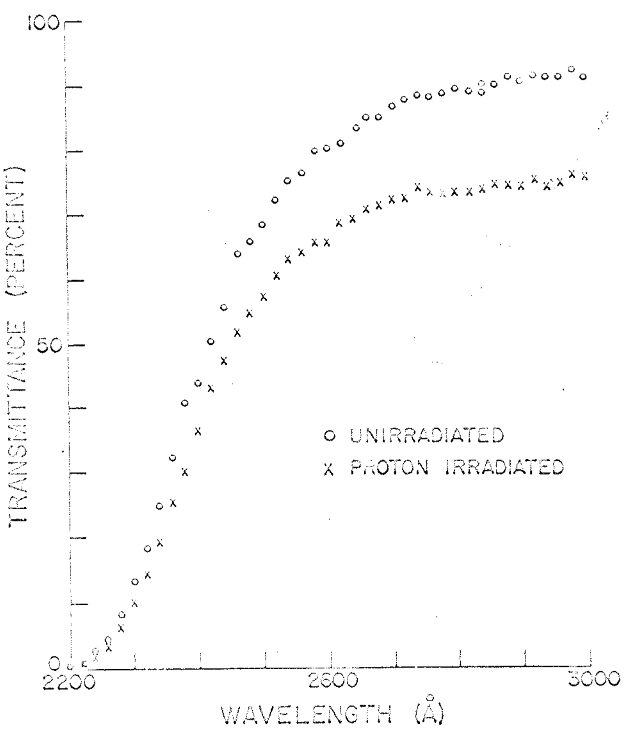

Figure 13—Transmittance of Corning 9-54 before and after irradiation by a total flux of 10^{12} protons/cm² at 5 MeV.

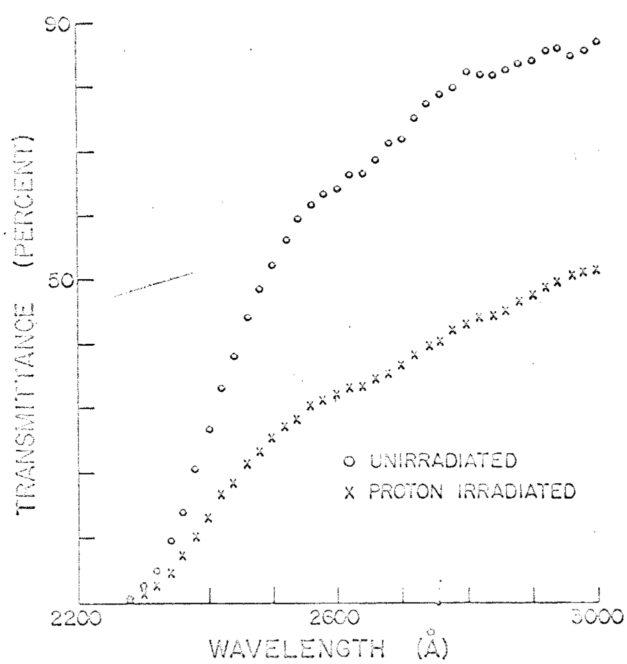

Figure 14—Transmittance of Corning 7-54 before and after irradiation by a total flux of 10^{12} protons/cm² at 5 MeV.

environment on a group of optical materials which are commonly used in the ultraviolet between 1050 Å and 3000 Å. These materials were all subjected to the same radiation dose and therefore it is possible to compare their transmittances and determine which are most suitable for use in a proton environment.

Of the materials listed in Table I, only LiF, NaF, NaCl, Corning 9-54, and Corning 7-54 were affected by the proton irradiation. A previous study by Heath and Sacher on the ultraviolet transmittances of optical material under electron irradiation indicated that LiF, Corning 9-54, and Corning 7-54 were damaged by the electron irradiation.[1] In their study, however, the Corning 9-54 and Corning 7-54 were shielded during irradiation by a thick synthetic sapphire crystal. This previous study of the transmittances under electron irradiation did not examine the transmission properties of NaF and NaCl.

It is apparent from the transmission curves that proton irradiation is not as damaging as electron irradiation on the transmittances of the optical materials studied. If care is taken to shield these optical components from electron irradiation in space, the shielding should also be effective against proton irradiation.

VI. ACKNOWLEDGMENTS

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