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Radiation Effects in Polymers for Plastic Scintillation Detectors

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Abstract

Radiation damage studies were performed on polystyrene and poly(vinyltoluene) samples containing different concentrations of either an antioxidant (AO-2) or a plasticizer (PP-4). In addition, parallel studies were carried out utilizing samples of these polymers prepared in the presence of cross-linking agents such as NPG, HDA, and DVB. The samples were irradiated using a ⁶⁰Co source to total doses of 1 and 10 Mrad, at a dose rate of approximately 1 Mrad/h. Transmittance measurements were recorded before and immediately after irradiation, and after oxygen annealing. These experiments showed that none of these agents improved the radiation resistance of polystyrene and poly(vinyltoluene) with regard to their optical properties.

Introduction

Plastic scintillators, which consist of a polymer matrix doped with fluorescent compounds (dopants), are noteworthy for their fast response. In addition, their relatively low cost and ease of manufacture facilitates their use in numerous detector geometries. Recently, new applications for this type of detector have resulted from developments in both scintillating plastic optical fibers and photon detection devices [1]. In fact, their utilization in future experiments may only be limited by their resistance to radiation damage. The results reported here are part of an ongoing project that focuses on the investigation and development of radiation resistant plastic scintillators.

Various studies have shown that the light yield in plastic scintillators decreases as their integrated exposure to ionizing radiation increases [2]. This effect could be caused by either polymer or dopant degradation. Further research has indicated that most of the commonly used fluorescent compounds do not degrade under radiation, but that the polymer does [3, 4]. Transmittance measurements of undoped polystyrene and poly(vinyltoluene) samples irradiated at high dose rate show a large absorption increase immediately after irradiation. A fraction of this absorption is transient in nature and will disappear with time. This annealing process can be enhanced by treating the samples with oxygen or heating them under vacuum. A permanent absorption remains as an indication of the radiation-induced changes that occurred in the molecular structure. The radiation-induced damage in the polymer is more significant in the blue/violet spectral region where most standard plastic scintillators emit. Thus, the light emitted by the dopants is partially reabsorbed by the damaged polymer. One approach to reduce such light output losses is to use fluorescent compounds which emit in the green/yellow region of the visible spectrum where changes in polymer transmittance after irradiation are small. Other possibilities include improvement of the polymer radiation resistance using additives to stabilize the material, or the utilization of new polymers other than polystyrene and poly(vinyltoluene) [5].

This study compares the radiation-induced damage in samples of pure polystyrene and poly(vinyltoluene) to those samples containing additives such as antioxidants, plasticizers, and cross-linking agents.

Experimental Section

Styrene and 4-methylstyrene were first deinhibited and then purified by vacuum distillation. The glass polymerization tubes were cleaned and treated with a solution to enable the removal of the plastic rods after polymerization. The monomer and the corresponding additive were placed in the test tubes, degassed with repeated freeze-pump-thaw cycles, and polymerized in a silicone oil bath without the presence of an initiator. The polymerized samples were cut into 1-cm thick discs and then polished. The antioxidant (AO-2) and the plasticizer (PP-4) were used at a 0.3% (by weight) concentration. The cross-linking agents, neopentyl glycol dimethacrylate (NPG) and 1,6-hexanediol diacrylate (HDA), were utilized at a 0.5% (by weight) concentration, but the concentrations for divinylbenzene (DVB) were of 0.01% and 20% (by weight). Transmittance spectra were recorded using a Hewlett-Packard 8451A spectrophotometer. An identical, unirradiated sample was used as reference. The irradiations were performed at Phoenix Memorial Laboratory (University of Michigan) using a ^{60}Co source. In all exposures, the dose rate was approximately 1 Mrad/h, and the total doses were 1 and 10 Mrad. The samples were irradiated under nitrogen atmosphere.

Results and Discussion

Transmittance measurements were recorded before irradiation, immediately after irradiation, and after an oxygen annealing period. In order to assess the losses due to radiation-induced damage, the transmittance spectra were integrated at each stage from 330 nm to 750 nm. The area of the absorption band measured immediately after irradiation or after the annealing process was subtracted from the transmission range available before irradiation. Various sample sets were prepared based on different polymers. The polymer matrices considered were polystyrene (PS), poly(vinyltoluene) (PVT), and a polystyrene/poly(vinyltoluene) copolymer (PS/PVT). Each set included one sample of (a) pure polymer as a reference, (b) polymer plus AO-2, (c) polymer plus PP-4, (d) polymer plus NPG. In addition, samples with DVB and HDA were prepared for the PS series. The selection of AO-2 and PP-4 was based on their performance in commercial plastics where their use improves the optical properties of the materials under radiation exposure.

However, these additives had not been tested for PS and PVT. Figures 1–4 show representative transmittance spectra of the cases studied.

In the PS set, with the exception of the samples doped with AO-2, all samples exhibited full transmission in the 330–750 nm range. Immediately after irradiation, they presented a large absorption band with a shoulder at 470 nm and a small band at 525 nm. The losses in the transmittance spectra at this stage were approximately 14% after 1 Mrad and 28% after 10 Mrad, and were more severe in the 330–550 nm range. After annealing, a residual absorption with a cut-off at approximately 480 nm remains. The permanent losses in the transmittance spectra are about 9% for 1 Mrad and 14% for 10 Mrad. The samples containing AO-2 presented an absorption band up to 380 nm before irradiation. The addition of the antioxidant altered the optical quality of the sample. Its transmittance spectra were poorer at all three stages that were measured.

In the PVT series, the results are similar to those of the PS sample set with the exception of the samples doped with AO-2. The presence of AO-2 in PVT samples did not increase their absorption. All samples exhibited full transmission between 330–750 nm. The area of the radiation-induced absorptions was integrated to calculate the losses from the initial transmission range. The losses in the transmittance spectra were approximately 11% after 1 Mrad and 29% after 10 Mrad; the permanent losses are about 8% and 14% respectively, after annealing.

In PS/PVT series, the results are similar to those of the PVT series. Before irradiation, the samples showed full transmission between 330–750 nm. The losses in the transmittance spectra were calculated to be approximately 11% after 1 Mrad and 27% after 10 Mrad; the permanent losses are about 8% and 14% respectively, after annealing.

Conclusions

The goal was to investigate if the presence of additives (antioxidants or plasticizers) or the cross-linked nature of the polymer would improve the optical quality of polystyrene or poly(vinyltoluene) under radiation. The results indicate that, in both cases, the polymer still shows an increase in its absorption after exposure to radiation. The additives studied do not improve the radiation resistance of these polymers under the conditions tested.

Acknowledgements

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References

- [1] *Proceedings of the Workshop on Scintillating Fiber Detectors*, edited by R.C. Ruchti, M. Wayne, and A.D. Bross (World Scientific, Singapore, to be published).
- [2] S. Majewski and C. Zorn, "Fast Scintillators for High Radiation Levels", in *Instrumentation in High Energy Physics*, F. Sauli, Ed., World Scientific, Singapore, 1992, pp. 157-279.
- [3] R.L. Clough *et al.*, *Proceedings of the Workshop on Radiation Hardness of Plastic Scintillators*, Florida State University, Tallahassee, FL, pp. 15-17 (1990).
- [4] A. D. Bross and A. Pla-Dalmau, *ACS Symposium Series 475*, 578 (1991).
- [5] X. Llop *et al.*, *Nucl. Instr. Meth.*, to be published.

Figure Captions

Figure 1. Transmittance spectra of a pure PVT sample.

Figure 2. Transmittance spectra of a PVT + 0.3% AO-2 sample.

Figure 3. Transmittance spectra of a PS + 0.3% PP-4 sample.

Figure 4. Transmittance spectra of a PS/PVT + 0.5% NPG sample.

3V0 - V SERIES - PVT+

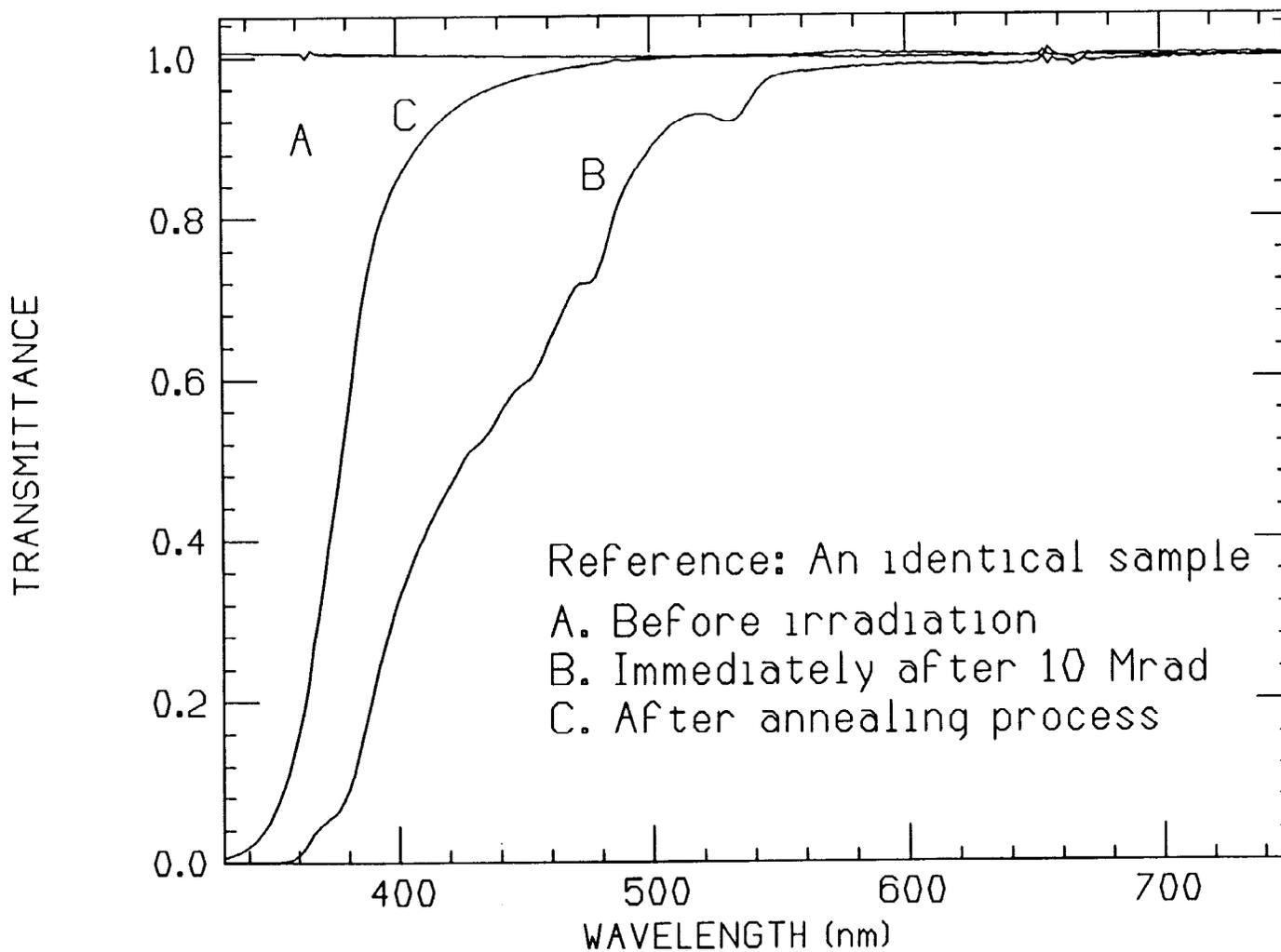


Figure 1

3V1 - V SERIES - PVT+

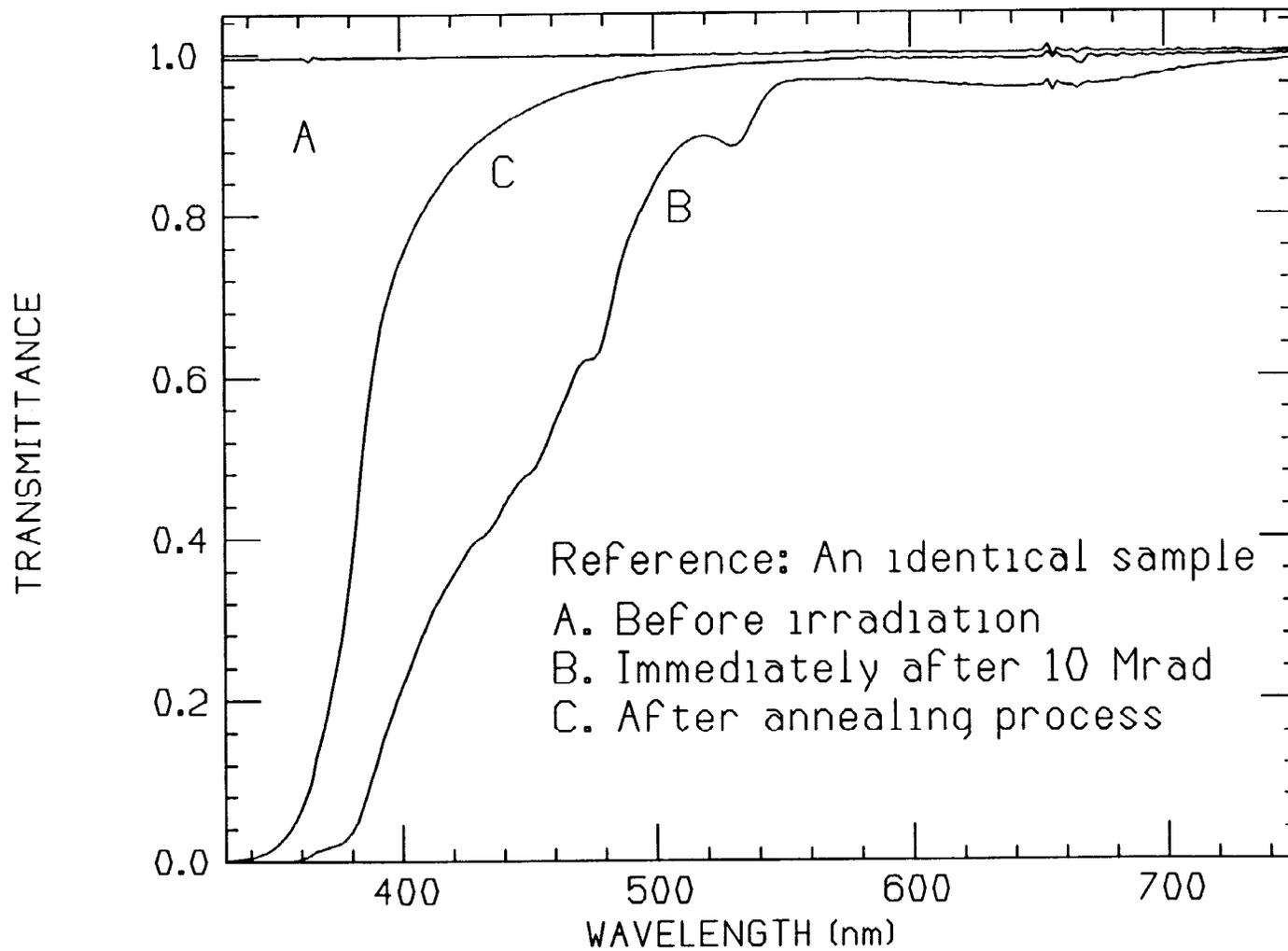


Figure 2

2S2 - S SERIES - PS+

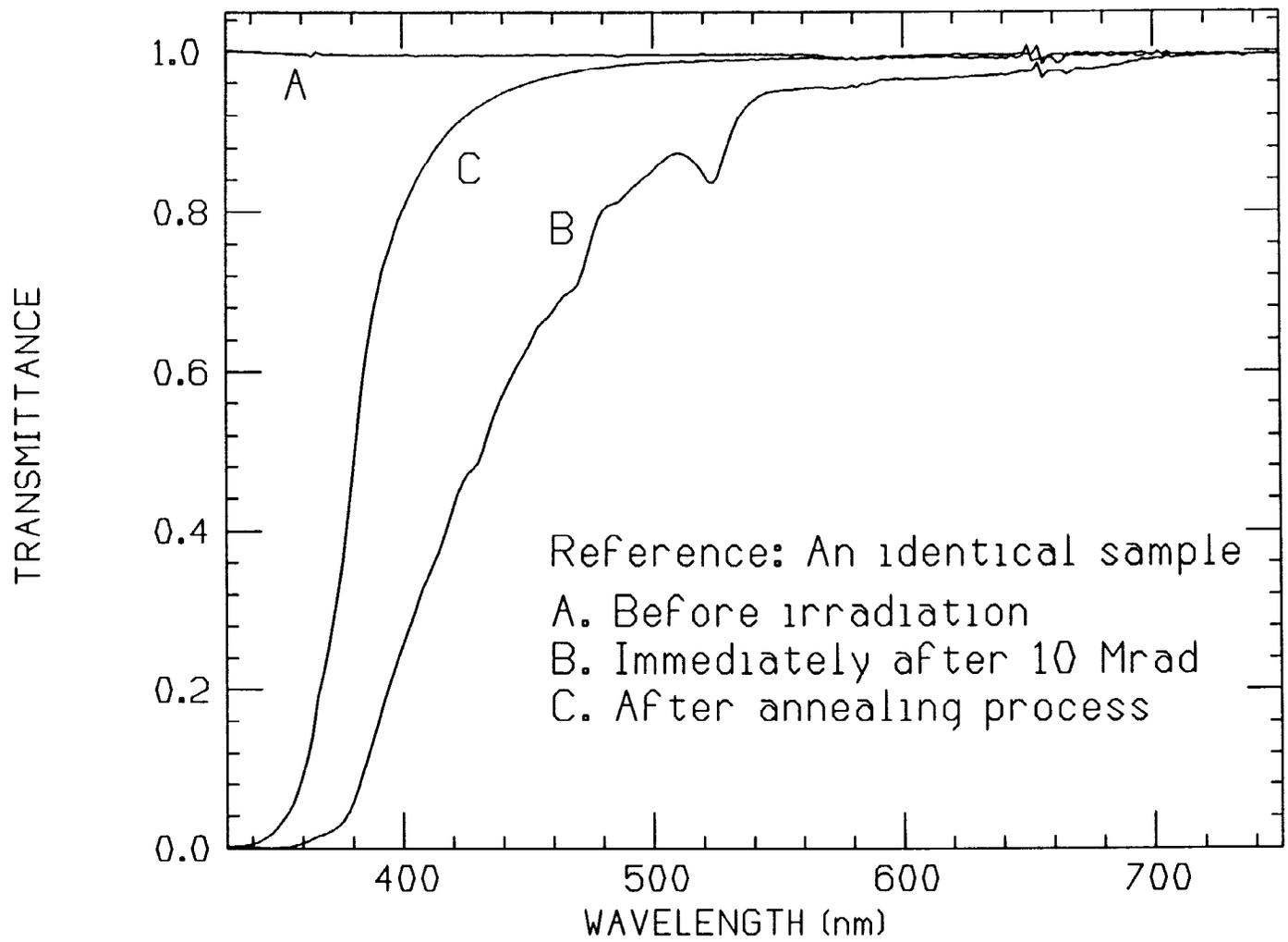


Figure 3

3R3 - R SERIES - POLYSTYRENE COPOLYMER

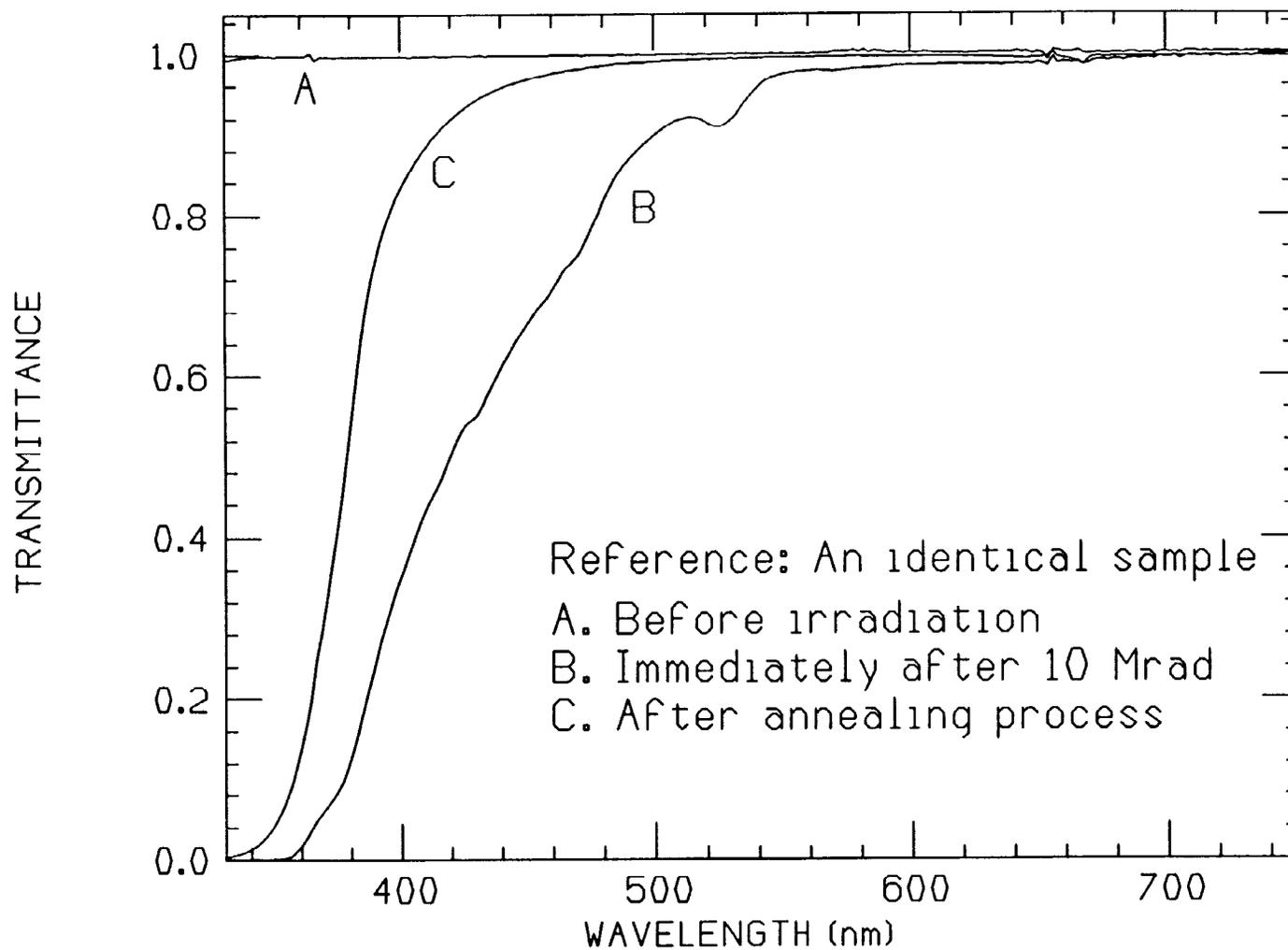


Figure 4