

KFK-222

**KERNFORSCHUNGSZENTRUM  
KARLSRUHE**

Mai 1964

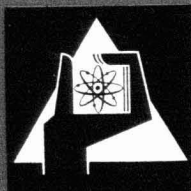
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Institut für Strahlenbiologie

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of Polyethylene Terephthalate

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Reprint from  
"BIOLOGICAL EFFECTS  
OF NEUTRON AND PROTON IRRADIATIONS"  
Vol. I

Gesellschaft für Kernforschung m.b.H.  
Zentralbücherei 1. Juli 1964

INTERNATIONAL ATOMIC ENERGY AGENCY  
VIENNA, 1964

<b>Büroexemplar</b>	Nr.
Gesellschaft für Kernforschung m.b.H. Karlsruhe	2

# SLOW-PROTON IRRADIATION OF POLYETHYLENE TEREPHTHALATE

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Abstract — Résumé — Аннотация — Resumen

SLOW PROTON IRRADIATION OF POLYETHYLENE TEREPHTHALATE. Epithermal neutrons interact with matter producing recoil protons which in turn transfer energy by ionization and by collision with nuclei. In an attempt to separate the effects produced by these various processes foils of polyethylene terephthalate (Hostaphan, Melinex or Mylar) were irradiated with slow protons in the energy range from 2 to 12 keV and the changes of the optical absorption caused by the irradiation measured spectrophotometrically.

Densely ionizing radiations (slow protons, 3.4 MeV alpha particles, and 2 MeV protons) induce in Hostaphan a sharp absorption maximum at 308  $\mu$ . At high doses of slow protons this maximum reaches a saturation value. The height of this saturation value increases with increasing proton energy. The  $D_{37}$  (1. e. dose to reach 63% of the saturation value) for protons from 2 to 12 keV is  $4 \times 10^{14}$  proton/cm<sup>2</sup> ( $\pm 20\%$ ).

To produce the maximum at 308  $\mu$  slow protons are more efficient than 3.4 MeV alpha particles (by a factor of 1.2 to 1.7) and 2 MeV protons (by a factor of 1.7). The reason might be that slow protons with velocities appreciably smaller than  $v_0$  ( $v_0 = c/137 =$  velocity of an electron in the lowest orbit of a hydrogen atom) ionize with a higher probability than generally assumed.

Irradiation with low energy protons produces also a broad absorption maximum at 250  $\mu$ , which is not observed after irradiation with 2 MeV protons (in vacuum) or with 3.4 MeV alpha particles (in helium under atmospheric pressure). At higher doses this maximum also reaches a saturation value, but this value is equal for all proton energies between 2 and 12 keV and corresponds to a  $D_{37}$  of  $0.8 \times 10^{14}$  proton/cm<sup>2</sup>. This maximum is thought to be due to a mechanism which has an appreciable efficiency only below 2 keV proton energy. The nature of this mechanism is unknown, but it may be caused either by a chemical reaction of the zero energy protons with the plastic molecules or by elastic collisions of the protons with atomic nuclei. It is remarkable, however, that this mechanism (as evident from the  $D_{37}$  values) is more efficient in changing the properties of Hostaphan than the ionization mechanism of densely ionizing radiation (slow protons or 2 MeV protons, respectively).

The experiments described here show clearly that the energy of very slow protons which are produced by epithermal neutrons is transferred to matter not only by ionization processes but also by another mechanism occurring at low proton energies. This mechanism of energy transfer, if shown to lead to biologically important consequences may have a bearing on setting tolerance limits for epithermal neutrons.

IRRADIATION DU TÉRÉPHTHALATE DE POLYÉTHYLÈNE PAR DES PROTONS LENTS. L'interaction des neutrons épithermiques et de la matière produit des protons de recul qui, à leur tour, transfèrent de l'énergie par ionisation et par collision avec les noyaux. Pour essayer de séparer les effets de ces divers processus, l'auteur a irradié des feuilles de téréphtalate de polyéthylène (Hostaphan, Melinex ou Mylar) avec des protons lents de 2 à 12 keV et il a mesuré par spectrophotométrie les modifications de l'absorption optique causées par l'irradiation.

Les rayonnements fortement ionisants (protons lents, particules alpha de 3,4 MeV et protons de 2 MeV) induisent dans l'Hostaphan une intense absorption, qui est maximum à 308  $\mu$ . A des doses élevées de protons lents, ce maximum atteint une valeur de saturation. Cette valeur de saturation augmente avec l'énergie des protons. La  $D_{37}$  (c'est-à-dire, la dose nécessaire pour atteindre 63% de la valeur de saturation) pour les protons de 2 à 12 keV est  $4 \cdot 10^{14}$  protons/cm<sup>2</sup> ( $\pm 20\%$ ).

Pour obtenir ce maximum à 308  $\mu$ , les protons lents sont plus efficaces que les particules alpha de 3,4 MeV (de 1,2 à 1,7 fois) et que les protons de 2 MeV (1,7 fois). On peut en voir la raison dans le fait que pour des protons lents ayant des vitesses notablement inférieures à  $v_0$  ( $v_0 = c/137 =$  vitesse de l'électron d'un atome d'hydrogène), la probabilité d'ionisation est plus grande qu'on ne le pense généralement.

L'irradiation par des protons de basse énergie produit également un large maximum d'absorption à 250  $\mu$ m, que l'on n'observe pas après l'irradiation par des protons de 2 MeV (dans le vide) ou par des particules alpha de 3,4 MeV (dans l'hélium à la pression atmosphérique). A des doses plus élevées, ce maximum atteint également une valeur de saturation, qui est la même pour toutes les énergies des protons entre 2 et 12 keV et correspond à une  $D_{37}$  de  $0,8 \cdot 10^{14}$  protons/cm<sup>2</sup>. On pense que ce maximum est dû à un mécanisme qui n'a une efficacité appréciable qu'au-dessous d'une énergie de 2 keV. On ne connaît pas la nature de ce mécanisme, mais il peut être causé soit par une réaction chimique des protons d'énergie zéro avec les molécules plastiques, soit par des collisions élastiques des protons avec des noyaux atomiques. Il convient toutefois de noter que ce mécanisme (tel qu'il apparaît d'après les valeurs de la  $D_{37}$ ) modifie plus efficacement les propriétés de l'Hostaphan que le mécanisme d'ionisation des rayonnements fortement ionisants (protons lents ou protons de 2 MeV, selon le cas).

Les expériences décrites dans le mémoire montrent clairement que l'énergie des protons très lents, tels qu'ils sont produits par les neutrons épithermiques, est transférée à la matière, non seulement par des processus d'ionisation, mais également par un autre mécanisme se produisant aux basses énergies protoniques. Ce mécanisme de transfert d'énergie peut avoir une influence sur l'établissement des limites de tolérance pour les neutrons épithermiques, si l'on établit qu'il a des conséquences importantes du point de vue biologique.

**ОБЛУЧЕНИЕ МЕДЛЕННЫМИ ПРОТОНАМИ ПОЛИЭТИЛЕНОВОГО ТЕРЕФТАЛАТА.**  
Надтепловые нейтроны взаимодействуют с веществом, производя протоны отдачи, которые в свою очередь передают энергию путем ионизации и столкновений с ядрами. Для разделения эффектов, производимых этими различными процессами, фольгу из полиэтиленового терефталата (Хостафан, Мелинекс или Милар) облучали медленными протонами с энергией от 2 до 12 кэв в спектрофотометрически измеряли изменения оптической абсорбции, вызванной облучением.

Излучения с высокой плотностью ионизации (медленные протоны, альфа-частицы с энергией 3,4 Мэв и протоны с энергией 2 Мэв) вызывают в хостафане резкий максимум абсорбции при 308 мкм. При высоких дозах медленных протонов этот максимум достигает величины насыщения. Высота этой величины насыщения возрастает с увеличением энергии протонов.  $D_{37}$  (т. е. доза, необходимая для достижения 63% величины насыщения) для протонов с энергией от 2 до 12 кэв равняется  $4 \cdot 10^{14}$  п/см<sup>2</sup> ( $\pm 20\%$ ).

Для получения максимума при 308 мкм медленные протоны являются более эффективными, чем альфа-частицы с энергией 3,4 Мэв (на фактор от 1,2 до 1,7) и протоны с энергией 2 Мэв (на фактор 1,7). Причина может состоять в том, что медленные протоны со скоростями значительно меньшими, чем  $v_0$  ( $v_0 = c/137 =$  скорости электрона, вращающегося по наименьшей орбите атома водорода) ионизируют с большей степенью вероятности, чем обычно предполагали.

Облучение протонами низких энергий вызывает также широкую максимальную абсорбцию при 250 мкм, которой не наблюдается после облучения протонами с энергией 2 Мэв (в вакууме) или альфа-частицами с энергией 3,4 Мэв (в гелии при атмосферном давлении). При более высоких дозах этот максимум достигает также величины насыщения, но эта величина одинакова для всех энергий протонов в диапазоне от 2 до 12 кэв и соответствует  $D_{37}$   $0,8 \cdot 10^{14}$  п/см<sup>2</sup>. Этот максимум, как полагают, должен быть связан с механизмом, который имеет заметную эффективность только для протонов с энергией ниже 2 кэв. Характер этого механизма неизвестен, но он может вызываться или химической реакцией протонов нулевой энергии с пластичными молекулами или упругими столкновениями протонов с атомными ядрами. Примечательно, однако, что этот механизм (как видно из величин  $D_{37}$ ) является более эффективным для изменения свойств хостафана, чем ионизационный механизм радиации с высокой плотностью ионизации (медленные протоны или протоны с энергией 2 Мэв соответственно).

Описанные здесь эксперименты ясно показывают, что энергия очень медленных протонов, производимых нейтронами, передается веществу не только при процессах ионизации, но и с помощью другого механизма, действующего при малых энергиях протонов. Этот механизм передачи энергии, который, как показано, ведет к биологически важным последствиям, может иметь значение при установлении пределов устойчивости адельфовых нейтронов.

**IRRADIACIÓN DEL TEREFTALATO DE POLIETILENO CON PROTONES LENTOS.** Por interacción de los neutrones epitérmicos con la materia, se producen protones de retroceso que a su vez transmiten energía por ionización y por cheque con los núcleos. A fin de diferenciar los efectos debidos a estos distintos procesos, el autor irradió láminas de tereftalato de polietileno (Hostaphan, Melinex o Mylar) con protones lentos de energías

comprendidas entre 2 y 12 keV y midió espectrofotométricamente las variaciones de su absorción óptica provocadas por la irradiación.

Las radiaciones de elevada densidad de ionización (protones lentos, partículas alfa de 3,4 MeV y protones de 2 MeV) provocan en el Hostaphan un pronunciado máximo de absorción a 308  $\mu$ . Para dosis elevadas de protones lentos, este máximo alcanza un valor de saturación. La magnitud de dicho valor de saturación crece al aumentar la energía de los protones. La  $D_{37}$  (es decir, la dosis necesaria para alcanzar el 63% del valor de saturación) para los protones de 2 a 12 keV es  $4 \cdot 10^{14}$  protones/cm<sup>2</sup> ( $\pm 20\%$ ).

Por lo que respecta al máximo en 308  $\mu$ , los protones lentos son de 1,2 a 1,7 veces más eficaces que las partículas alfa de 3,4 MeV y 1,7 veces más eficaces que los protones de 2 MeV. Ello se explica al parecer por el hecho de que los protones lentos, de velocidades apreciablemente inferiores a  $v_0$  ( $v_0 = c/137 =$  velocidad de un electrón en la órbita más baja de un átomo de hidrógeno) ejercen una acción ionizante con un grado de probabilidad mayor que el generalmente admitido.

La irradiación con protones de baja energía también produce un amplio máximo de absorción a 250  $\mu$ , que no se observa después del bombardeo con protones de 2 MeV (en el vacío) ni con partículas alfa de 3,4 MeV (en helio a presión atmosférica). Con dosis mayores este máximo alcanza también un valor de saturación, pero en este caso dicho valor permanece constante para todas las energías protónicas comprendidas entre 2 y 12 keV, y corresponde a una  $D_{37}$  de  $0,8 \cdot 10^{14}$  protones/cm<sup>2</sup>. Se supone que este máximo se debe a un mecanismo cuya eficiencia sólo se hace apreciable por debajo de una energía protónica de 2 keV. La naturaleza de ese mecanismo no se conoce, pero es posible que sea causado por una reacción química entre los protones de energía cero y las moléculas del material plástico o bien por los choques elásticos entre los protones y los núcleos atómicos. De todos modos, es notable que dicho mecanismo (según se desprende de los valores de  $D_{37}$ ) sea más eficiente para alterar las propiedades del Hostaphan, que la ionización de las partículas densamente ionizantes (protones lentos o protones de 2 MeV, respectivamente).

Los experimentos descritos en la memoria demuestran claramente que la energía de los protones muy lentos, tales como los que producen los neutrones epitérmicos, es transmitida a la materia no solamente en virtud de procesos de ionización, sino también por otro mecanismo que interviene a energías protónicas bajas. Si se comprobara que este mecanismo de transmisión de energía tiene consecuencias biológicas importantes, podría influir en la imposición de límites de tolerancia para los neutrones epitérmicos.

## INTRODUCTION

It is generally assumed that the tolerance dose for epithermal neutrons lies between the tolerances for thermal and for fast neutrons. By comparing the way thermal, epithermal, and fast neutrons interact with matter such an assumption does not seem to be justified without further experimental proof [1 - 4].

To calculate the dose for thermal neutrons the well-known cross-sections for neutron capture are used. In biological material two important reactions are possible: radiative capture by hydrogen and an (n, p) process with nitrogen. These reactions produce 2.2-MeV gamma rays and 660-keV protons, respectively. From the ionization efficiency of these radiations the energy absorbed by the material is calculated. Fast neutrons lose energy mainly by elastic collisions leading to recoil protons. When slowed down to thermal energies the neutrons undergo capture reactions as set out above. From the ionization efficiency of the recoil protons and that of the radiations produced in nuclear reactions the dose for fast neutrons is calculated.

The dose for epithermal neutrons has usually been determined in the same way. The recoil protons liberated in elastic collisions have a small cross-section for ionization (according to theory). When the dose is calculated from the ionizations only the proportion of the dose delivered by

the recoil protons is about 1/10 of the total dose. The major fraction is supplied by the nuclear reactions of the neutrons slowed down to thermal energies [5]. The small contribution by recoil protons to the total dose is somewhat surprising. In material containing hydrogen a slow neutron of e. g. 1 keV energy can knock several hydrogen atoms out of their molecular positions until the energy of the primary neutron is so reduced that this process is no longer possible. The slow recoil protons thus produced can liberate more protons by elastic collisions. Together with these secondary recoil protons the total number of hydrogen atoms removed from their molecules is considerable. It is, however, not possible to estimate the total number of recoil protons produced in this way as it is unknown which part of the energy of the recoil protons is used for the excitation of vibrational and rotational levels of the molecules and which part of the energy is transferred in the process of electron capture and loss. This last mechanism certainly occurs at very low energies but it is usually ignored in dose calculations. Furthermore it is possible that the recoil protons after having lost their kinetic energy react with other hydrogen atoms. By this mechanism the number of the hydrogen atoms removed from their molecules can be additionally increased without the occurrence of ionization. Finally, radiation energy is transferred to matter by the removal of a hydrogen atom from a molecule and not only by the removal of an electron.

The above considerations of elementary processes justify the question why in most estimations of dose for slow particles the energy absorbed in electronic processes only is taken into account, whereas the energy transferred in elastic collisions has usually been neglected. They also indicate the need for detailed experimental studies of which the present paper forms a part.

## MATERIAL AND METHODS

Up to now no experiments have come to our knowledge dealing with the biological effects of epithermal neutrons. Probably this is due to experimental difficulties in the production of monoenergetic slow neutrons (at high intensities) and in the dosimetry of these neutrons. Moreover, in such experiments one would always have to deal with a mixture of effects due to recoil protons produced by the neutrons and of effects caused by radiations arising from nuclear reactions. It is, however, possible to separate these two effects by irradiations with low-energy protons. As a simple system for such experiments plastic foils can be used. It was first reported in 1951 that the absorption spectrum of plastic foils is changed by ionizing radiations [6]. But it is difficult to determine which changes within the plastic material are responsible for the observed changes in the optical transmission. One would expect specific reactions like production of stable radicals, main chain breaks, or cross-linking to cause characteristic changes in absorption. Consequently, one may hope to differentiate between different mechanisms of energy transfer leading to different reactions by the appearance of different absorption spectra.

To verify this assumption irradiation experiments were carried out with thin foils of polyethylene terephthalate commercially obtainable as

Hostaphan\* and equivalent to Melinex and Mylar. In the experiments foils of  $6.5\ \mu\text{m}$  thickness were used – the thinnest foils readily available. Hostaphan contains no plasticizers or polymerizers and has a constant chemical composition. This fact is most important, since in experiments with particles of only  $200\ \text{\AA}$  range a varying distribution of additional substances will easily cause experimental errors.

The absorption spectrum of an unirradiated  $6.5\text{-}\mu\text{m}$  thick Hostaphan foil is given in Fig. 1. The foil is transparent to visible light but its absorption increases rapidly at wave lengths below  $315\ \text{nm}$ . In order to determine small changes of transmission in the ultraviolet region the absorption of the foils was measured before and after irradiation in comparison to the same control foil in a spectrophotometer (Zeiss PMQ II). The difference of these two readings gives the change in optical absorption produced by the irradiation and is largely independent of errors due to reflection and scattering.

The foils were irradiated in vacuo with slow protons from an accelerator, the construction and operating conditions of which will be described elsewhere. The range of proton energy was 2 to 12 keV.

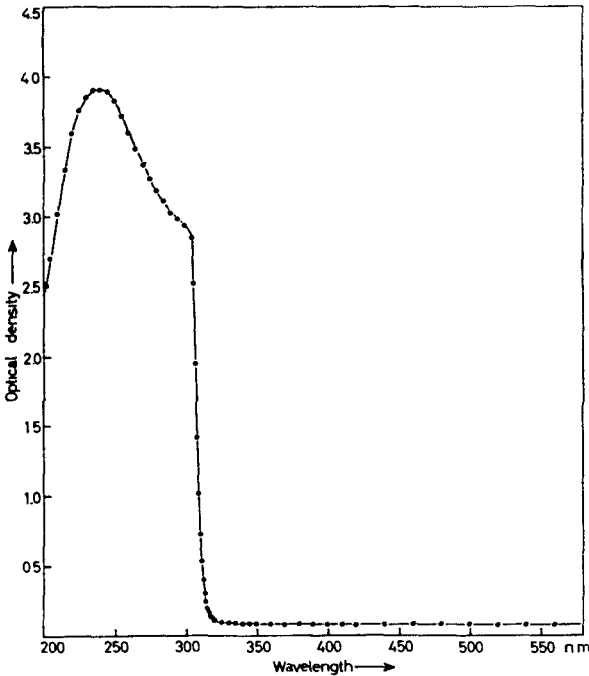


Fig. 1

Absorption spectrum of an unirradiated  $6.5\text{-}\mu\text{m}$  thick Hostaphan foil

\* Trade name: Kalle & Co., Wiesbaden, Germany.

## RESULTS

The absorption spectra produced in Hostaphan by irradiation with slow protons of different energies are shown in Fig. 2. The dose was about  $5 \times 10^{14}$  protons/cm<sup>2</sup> for all energies. A sharp maximum is observed at 308 nm, the height of which (compared with the absorption at 300 nm) increases with increasing proton energy. Beside this sharp peak there is a broad maximum at 250 nm. Its height (compared with the absorption at 300 nm) is equal for all proton energies. From this fact we may infer that the broad maximum at 250 nm is due to a mechanism which is efficient at proton energies below 2 keV.

In order to test this conclusion Hostaphan foils were irradiated in vacuo with 2-MeV protons from a high voltage Van de Graaff accelerator and also in helium under atmospheric pressure with 3.4-MeV alpha particles from

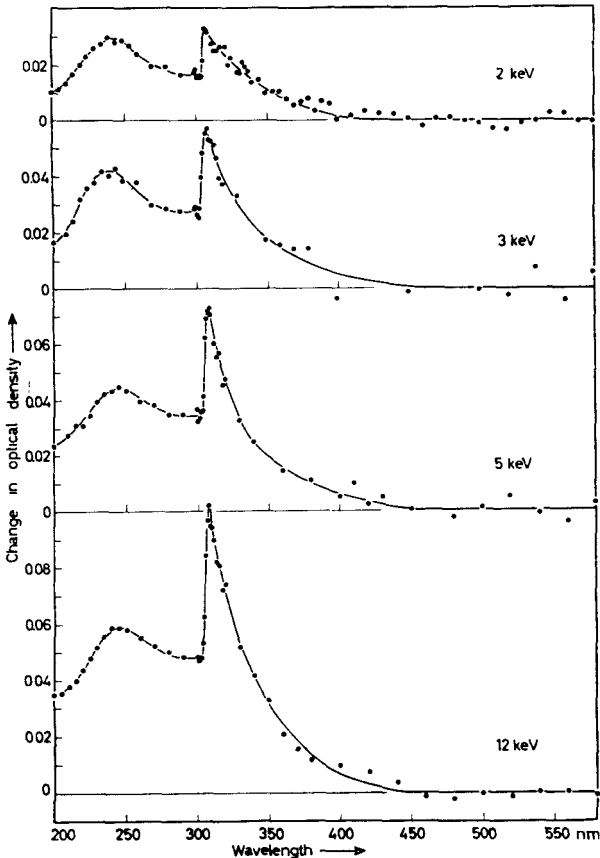


Fig. 2

Absorption spectra produced in Hostaphan by irradiation with slow protons of different energies, the doses being about  $5 \times 10^{14}$  protons/cm<sup>2</sup>



a  $\text{Po}^{210}$  source. In either case the particles have sufficient energy to traverse the foil completely losing about 100 keV and 800 keV, respectively. Therefore, mechanisms specific for the end of the range of the particles cannot contribute to the observed effect. The spectra thus obtained show the sharp peak at 308 nm but not the broad maximum at 250 nm (Fig. 3) demonstrating clearly that the absorption peak at 308 nm is caused by ionizing radiation.

The absorption peak at 308 nm measured for different proton energies increases with dose. Figure 4 shows for 12-keV protons that the height of this peak reaches a saturation value for high doses. This saturation can be explained by assuming that within the range of the protons all chemical bonds contributing to the peak at 308 nm have been altered. This assumption is supported by the fact that the logarithm of the difference between saturation value and change in optical density plotted against dose is a straight line (Fig. 5). Exactly the same result is found with proton energies of 2, 3, 5, and 8 keV. The saturation value, however, decreases with decreasing proton energy (i. e., with decreasing range of the protons). The  $D_{37}$  (dose to reach 63% of the saturation value) amounts to  $4 \times 10^{14}$  protons/cm<sup>2</sup> ( $\pm 20\%$ ) within the energy range studied, i. e., 2 to 12 keV. The initial slope of the dose-effect curves (Fig. 4) gives the efficiency to produce the peak at 308 nm. The energy expenditure for a change in optical density of 0.01 is 7.7 krad/cm<sup>2</sup> ( $\pm 20\%$ ) for slow protons and about 13 krad/cm<sup>2</sup> for 2-MeV protons. Slow protons are hereby more efficient in producing the peak at 308 nm than fast protons.

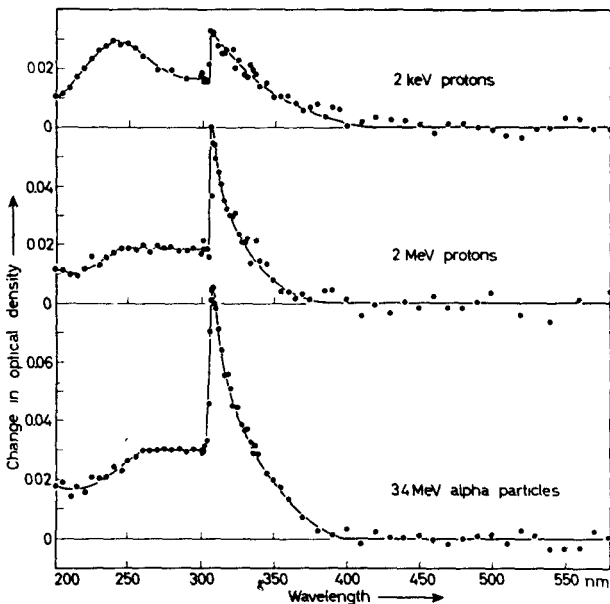


Fig. 3

Absorption spectra produced in Hostaphan by irradiation with 2-keV protons (13 krad), 2-MeV protons (45 krad) and 3.4-MeV alpha particles (about 75 krad)

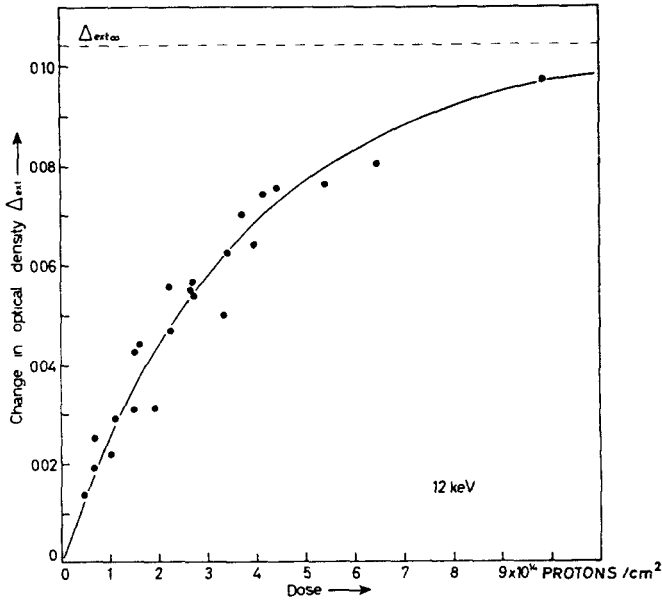


Fig. 4

Change in optical density at 308 nm produced in Hostaphan by irradiation with 12-keV protons at different doses

The increase of the broad absorption maximum at 250 nm plotted against dose of 2-keV protons reaches saturation value more quickly than the ionization peak at 308 nm (Fig. 6). The  $D_{37}$  for the increase of the 250 nm maximum derived from the half-logarithmic plot of the difference between saturation value and absorption change versus dose (Fig. 7) was determined to be  $0.8 \times 10^{14}$  protons/cm<sup>2</sup>. Comparing this dose to the dose values given above the broad absorption maximum is produced by a mechanism which is more efficient by a factor of 5 than the ionization or excitation mechanism of 2-keV protons and 8 times more efficient than the ionization or excitation mechanism of 2-MeV protons.

## DISCUSSION

It is not yet possible to say much about the kind of changes in the plastic material causing the observed spectra. Nevertheless, both maxima (at 308 nm and 250 nm) are obviously due to quite stable energy states. Storing in air at room temperature for eight weeks after irradiation or heating up to 100°C for one hour produced no appreciable change in the two maxima. The sharp peak at 308 nm is found after irradiation with 2-MeV protons or 3.4-MeV alpha particles as well as after irradiation with low-energy protons (2 to 12 keV). Therefore, it is evident that this peak is caused by

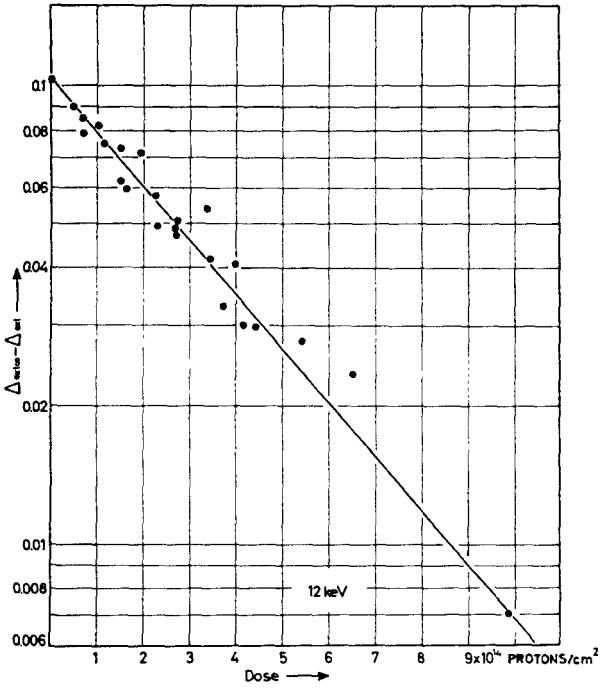


Fig. 5

Difference between saturation value ( $\Delta_{ext\infty}$ ) and change in optical density ( $\Delta_{ext}$ ) at 308 nm produced in Hostaphan by irradiation with 12-keV protons at different doses

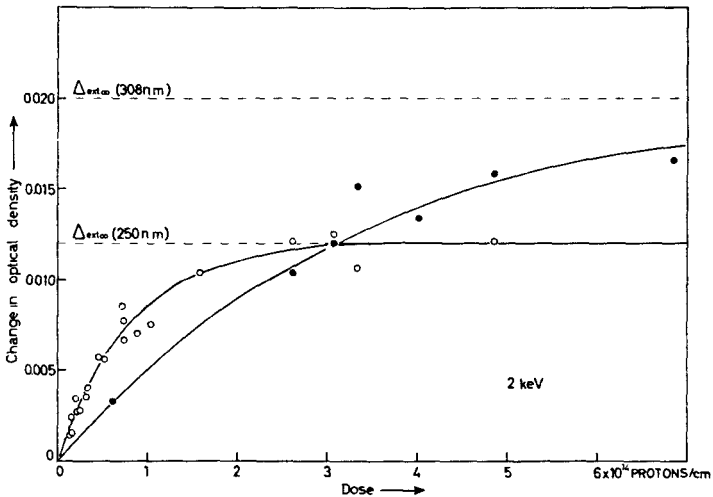


Fig. 6

Changes in optical density at 308 nm (black dots) and 250 nm (open circles) produced in Hostaphan by irradiation with 2-keV protons at different doses

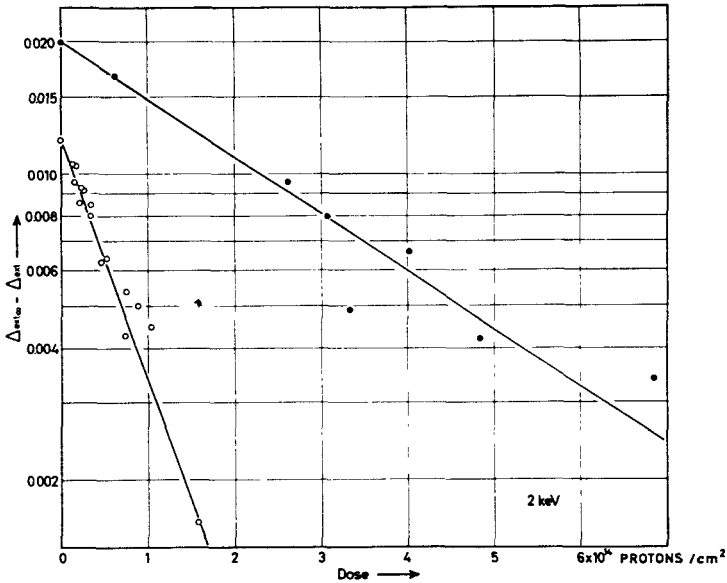


Fig. 7

Difference between saturation value ( $\Delta_{exte}$ ) and change in optical density ( $\Delta_{ext}$ ) at 308 nm (black dots) and 250 nm (open circles) produced in Hostaphan by irradiation with 2-keV protons at different doses

the ionization or excitation effect of the various radiations. It is remarkable, that slow protons are 1.7 times more efficient than 2-MeV protons to produce this ionization peak.

In the energy range from 2 to 12 keV the cross-section for electron capture is essentially higher than that for electron loss so that about 90% of the incident protons are neutral hydrogen atoms while passing through matter [7]. The ionization cross-section of neutral hydrogen atoms below 22 keV energy is higher than that for protons of the same energy (by a factor of almost 2 as measured in hydrogen for particle energies of 9 keV [8]). Ionizations are also produced by the process of electron capture and loss. Both these effects are generally neglected in dose calculations. From these facts we may conclude that the ionization yield of low-energy protons is higher than normally assumed and thus may explain the efficiency of slow protons to produce the ionization peak at 308 nm.

Two possible mechanisms of producing the broad absorption maximum at 250 nm seem to deserve consideration here.

(i) A simple chemical reaction of the protons with the molecules of the irradiated substance.

After having lost their kinetic energy the protons are, in fact, atomic hydrogen which is known to be rather reactive. Thus the protons might react at the end of their paths with the molecules of the Hostaphan causing the absorption maximum at 250 nm. We have no evidence for this reaction so far but it is energetically possible to occur: The bond energy of R-H,

where R is any organic molecular residue, is 95-100 kcal/mole, the bond energy of hydrogen H-H is 103.4 kcal/mole. An experiment to investigate the influence of atomic hydrogen on Hostaphan is now under way.

(ii) Elastic collisions of the protons with atomic nuclei.

It is known that the cross-section for elastic collisions rises with decreasing proton energy whilst that for ionization falls (below 60 keV proton energy). According to calculations by SNYDER and NEUFELD [9, 10] the cross-section for ionization of 2-keV protons in tissue is about twice that for elastic collisions. To a first approximation this ratio may be taken to be applicable to Hostaphan also. In the experiments with 2-keV protons the saturation value for the ionization peak (at 308 nm) was found to be 0.02 and that of the broad maximum (at 250 nm) to be 0.012. Assuming that the products formed by the two different mechanisms have the same extinction coefficient per mole (which might be true within a factor of 10) these experiments agree with the cited calculations. On the basis of our present knowledge elastic collisions cannot, therefore, be excluded as a possible mechanism producing the peak at 250 nm. A clear proof, however, for or against this point is not yet available. Nevertheless, our experiments demonstrate clearly that slow protons are quite efficient in changing the properties of matter. Moreover they show that at very low proton energies there exists a mechanism of interaction which is 5 to 8 times more efficient in changing the properties of Hostaphan than densely ionizing radiations. Should analogous experiments using biological materials show a similar ratio of efficiencies for damage, tolerance doses for epithermal neutrons might have to be revised.

#### ACKNOWLEDGEMENTS

The author is most grateful to Professor K.G. Zimmer for his continued support and interest in this study. His thanks are due also to Dr.G. AHNSTRÖM and Professor L. EHRENBERG for kindly making available an unpublished report [11].

#### REFERENCES

- [1] MAGEE, J.L., KAMEN, M.D. and PLATZMAN, R.L., Basic mechanisms in radiobiology, Washington (1953).
- [2] FANO, U., "Principles of radiological physics", Radiation Biology (A. HOLLAENDER, Ed.) New York (1954).
- [3] ZIMMER, K.G., *Strahlentherapie* 101 (1956) 143-151.
- [4] RIEHL, N., *Atomkernenergie* 9 (1956) 297-300.
- [5] SCHAL'NOW, M.I., Tkanevaja dosa neutronov, Atomisdat, Moskva (1960); *Neutronengewebedosimetrie*, Deutscher Verlag der Wissenschaften, Berlin (1963).
- [6] DAY, M.I. and STEIN, G., *Nature* 168 (1951) 644-645.
- [7] STIER, P.M. and BARNETT, C.F., *Phys. Rev.* 103 (1956) 896-907.
- [8] SCHWIRTZKE, F., *Z. Physik* 157 (1960) 510-522.
- [9] SNYDER, W.S. and NEUFELD, J., *Rad. Res.* 6 (1957) 67-78.

- [10] NEUFELD, J. and SNYDER, W.S., "Estimates of Energy Dissipation by Heavy Charged Particles in Tissue", Selected topics in radiation dosimetry, IAEA, Vienna (1961) 35-44.
- [11] AHNSTRÖM, G., EHRENBERG, L. and ZIMMER, K.G., Report to statens råd för atomforskning, Stockholm (1960).

## DISCUSSION

J.F. FOWLER: Firstly, have you any information about the change in the optical absorption of Hostaphan as the result of irradiation by X- or  $\gamma$ -rays or fast electrons? Earlier work by Boag and others showed a peak outside the 250-nm region, which faded rapidly and which also depended upon the presence or absence of oxygen. Secondly, were your specimens irradiated in vacuo but stored in air?

H. JUNG: As you say, Boag and co-workers (J.W. Boag, G.W. Dolphin and J. Rotblat, Rad. Res. 9 (1958) 589), using high-energy electrons, found an absorption peak at 325 nm. Effects in the ultraviolet region were not measured, the spectra being recorded from 315 nm towards higher wave lengths. The peak at 325 nm consists of two components, one of which fades rapidly, the other remaining practically constant.

I have done some experiments using  $\text{Co}^{60}$   $\gamma$ -rays and the spectra obtained after irradiation under anaerobic conditions are the same as those found after irradiation with 2-MeV protons or 3.4-MeV alpha particles. Within the experimental accuracy of several per cent, no fading of the sharp absorption peak at 308 nm was found over a period of two months. Irradiation with  $\gamma$ -rays in the presence of oxygen induces absorption spectra of somewhat different shape. An additional maximum appears at 325 nm, which should be identical with the constant component reported by Boag, as the spectra were found to remain unchanged over many weeks. The dose rate used in my experiments with  $\gamma$ -rays was too low (about 1 mrad/h) to measure the fading component.

All proton experiments were performed in a high vacuum below  $10^{-5}$  mm Hg. After irradiation, the samples were stored in air, at room temperature.

G. AHNSTRÖM: Does the 308 nm peak move towards longer wave lengths as dose increases?

H. JUNG: We observed no shift of the maximum towards longer or shorter wave lengths in the dose range studied.

L.G. EHRENBERG: In studies of irradiated seeds, there are certain indications of qualitatively different effects - compared with those obtained with medium-energy neutrons - when there is a large contamination of epithermal neutrons, or at higher energies where a greater fraction of the dose is due to heavy ions (chiefly C and O). One such effect is a high sterility rate as compared with the mutation rate; another is a high frequency of small fragments - these are also observed as a consequence of  $\text{P}^{32}$  incorporation in the nuclear material, possibly a function of recoil  $\text{P}(\rightarrow\text{S})$  ions. These qualitative effects may be due to the types of mechanism discussed by Dr. Jung, or to high LET. Dr. Snyder has demonstrated the LET distribution of 0.1-MeV neutrons to be between those of 0.5- and 2.5-MeV neutrons. It would be of value to include in calculations of the kind made

by Dr. Snyder the fraction of energy dissipated in elastic collisions of particles.

H. JUNG: I agree, but this procedure is very difficult because of the entire lack of experimental data. It is not even possible to determine the absolute number of recoil protons liberated by a particle of a certain energy.

Professor Ehrenberg's experiments indicate that seeds have different responses to neutrons of different energies. If this is due to the occurrence of elastic collisions, it is not yet possible to treat this problem quantitatively, because we do not know the effectiveness of damage due to an elastic collision compared with that of an ionization. Much experimental work will therefore be necessary before the action of very slow particles is understood.