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THE USE OF ORGANIC MATERIAL MAKROCLEAR FOR RADIOCHROMIC INTEGRATING DOSIMETRY OF HADRON BEAMS

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Abstract. *The Laboratory of Cyclotrons and Fast Neutron Generators performed a series of experimental irradiations of MAKROCLEAR radiochromic integrating dosimeters by proton and deuteron beams accelerated on a U-120M cyclotron. These dosimeters have been developed at the Research Centre Rez.*
The dosimeters prepared in the form of small blocks were successively irradiated by protons and deuterons of various energies and in various doses. The results of the analyzes showed that MAKROCLEAR dosimeters are very useful as inexpensive and readily available integrating proton dosimeters in the dose range up to about 7.5 kGy, where their response in white light and monochromatic light with longer wavelenght (about 640 nm) is practically linear with a dose. An even higher measuring range was seen in the case of deuterons, where the response of dosimeters was linear with a dose up to 15 kGy.
Keywords: *Polymeric dosimeter, Mono & polychromatic transmission scanning, Optical density curves, Photometry, Bragg Curve, Optical Density, Hadron beams, Transmission scanner*

1. INTRODUCTION

1.1. The MACROCLEAR dosimeters

The MAKROCLEAR dosimeters have been developed at the Research Centre Rez at the Radiochemistry II department since 2016. It is a solid-state, clear organic polymeric material (Figure 1) that responds to irradiation by changes in optical density. For doses below 500 Gy, these changes first occur in the near UV region of the spectrum, then propagate into the light region and for doses above 1 MGy pass into the infrared region of the spectrum.

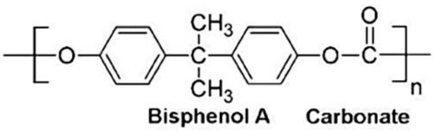

Figure 1. Polycarbonate (PC) - the basic structural unit of the organic material MAKROCLEAR

The dosimeters are evaluated photometrically, the simplest method being the scanning on an Epson Perfection 850-Pro transmission scanner with resolution up to 6400 x 9600 DPI with the possibility of using color filters that transmit only selected wavelengths of light [1]-[6].

Dosimetry of high doses of ionizing radiation is widely used not only in the field of basic and applied research, but also in various industrial sectors. For high dose dosimetry, the change in optical density is an easily and inexpensively measurable parameter. In this area, the use of solid-state organic radiochromic dosimeters is therefore much simpler than the use of alanine or gel dosimeters, which require very time-consuming and expensive laboratory methods for their evaluation (electron paramagnetic resonance or nuclear magnetic resonance).

Cited works [7]-[10] describe also the use of radiochromic films (gafchromik) for dosimetry of hadron beams. However, the range of measurable doses is different for gafchomic films – the maximum measurable dose is 1 kGy (HD-V2 film). The disadvantage of gafchromic films compared to clear MAKROCLEAR dosimeters is the rather high initial optical density (OD) of unirradiated films, which thus reduces the measurable range of optical densities after irradiation. Due to the planar nature of the gafchromic film, it is also necessary to usin a suitable phantom, between the plates of which the gafchromic film is inserted parallel to the axis of the beam. Air gaps between the film and the plates of the phantom can then cause significant artifacts in the measured percentage depth dose, as was described, for example, in [7]. The mentioned problems disappear with transparent 3D MAKROCLEAR dosimeters. The OD of non-irradiated polycarbonate is less than 0.1 cm-1, so it can be prepared in the desired volume without the need for an additional phantom, while still being much more transparent than a thin gafchromic foil.

The undisputed advantages of MAKROCLEAR dosimeters over other types include their easy preparation in the desired shape and size, their easy availability, and very low purchase price (approximately 1 Eurocent per piece). Their evaluation is also cheap, easy, fast and does not require expensive or bulky laboratory equipment. In a matter of minutes, 3D information about

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the dose profile of the hadron beam and the depth dose curve can be obtained, including the position of the Bragg peak and the dose at the Bragg peak, the ratio of the dose at the Bragg maximum to the dose in the plateau region, and the maximum particle range.

Since the dosimeter material has a density only 8% higher than the average density of the human body, the method can also be used in hadronic therapy of oncological diseases.

1.2. Interaction of heavy charged particles with the environment of the human body and with tissue equivalent materials

Conventional radiation oncology uses photon or electron beam irradiation, the source of which is usually linear accelerators. Most of the energy is transferred by these particles to the tissue lying on the surface of the body or just below it (Figure 2). As the depth of penetration into the tissue increases, the photons experience an exponential decrease in the transmitted energy after a sharp initial increase. Places in front of the target area can be irradiated by individual beams more than the actual bearing. Places behind the target area are also exposed to a certain radiation exposure. Irradiation from multiple fields can significantly reduce the impact of these adverse effects. For many tumors located in proximity of critical organs, conventional beam radiotherapy leads to a higher risk of unacceptable damage to these structures [11].

Heavy charged particles (protons, deuterons, tralphiones, alpha particles and other ions) lose their energy as they pass through the environment, mainly through collisions with electrons bound in the molecules of the irradiated environment. The interaction of the heavy charged particles with the environment is very well described using Bethe-Bloch theory. This theory describes not only energy losses, but also the lateral scattering of the particle and its range [11].

Accelerated heavy charged particles ionize atoms along their path as they pass through mass, thereby gradually losing their kinetic energy until they complete their path at a depth that can be calculated depending on the species and initial energy of the particle and the type of matter in which the energy is transferred. It is at the end of the path of the heavy charged particle that the ionization increases sharply, as the effective cross section for resonant capture of particles increases rapidly with decreasing velocity. In these places, the dose increases sharply. The depth dose curve (Bragg curve) thus shows the so-called Bragg peak at the end of the path [11].

The shape of the Bragg curve is the main principal advantage of heavy charged particles from the point of view of their therapeutic use. Heavy charged particles release a very small portion of their energy as they pass through the environment and transfer most of their energy just before the end of their trajectory. Therefore, the dose at the Bragg peak, where the target volume is located, is significantly greater than at the plateau area of the Bragg curve. In addition, the range of these particles is very well defined. Furthermore, in contrast to the particles used in conventional irradiation, the heavy charged particles hardly undergo lateral scattering, they pass through the tissue in a practically straight direction [11].

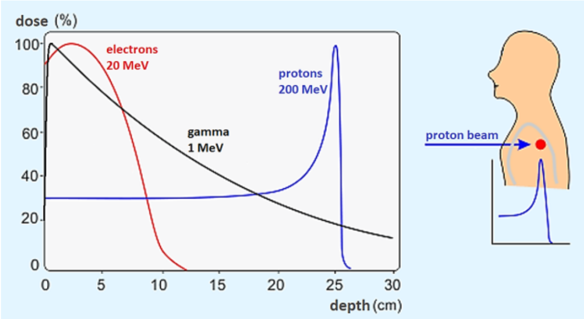

Figure 2: Left - the depth dose curves of photons 1 MeV (black), electrons 20 MeV (red), protons 200 MeV (blue). Right - the principle of using the Bragg peak to transport a high dose to the tumor site at depth. Source: [11].

1.3. The laboratory of Cyclotrons and Fast Neutron Generators

In 2021, the Laboratory of Cyclotrons and Fast Neutron Generators (LC & FNG) performed a series of experimental irradiations of MAKROCLEAR radiochromic integrating dosimeters by proton and deuteron beams accelerated on a U-120M cyclotron. LC & FNG is a part of the research infrastructure CANAM (Center of Accelerators and Nuclear Analytical Methods) of the Institute of Nuclear Physics of the Czech Academy of Sciences (Figure 3) [12].

Figure 3. Beam output from the U-120M cyclotron of Laboratory of cyclotrons and fast neutron generators of the Institute of Nuclear Physics AS CR

2. The course of the experiment

The dosimeters prepared in the shape of blocks of the dimensions 10 mm x 10 mm x 10 mm or 10 mm x 10 mm x 20 mm were placed with their longest dimension parallel to the axis of the hadron beam in a special aluminum holder, behind a 10 mm thick aluminum collimator with a 9 mm diameter aperture so that the axis of the proton beam passed through the center of the sample. (Figure 4 and 5).

The collimator with the sample was placed 500 mm in front of the exit window of the hadron beam from the accelerator.

Before irradiation, each hadron beam was pre-monitored by a Farmer ionization chamber equipped with a replaceable build-up layer specifically for each of the energies used. The ionization chamber was connected to a precision UNIDOS electrometer (PTW Freiburg) converting the collected charge into the absorbed dose.

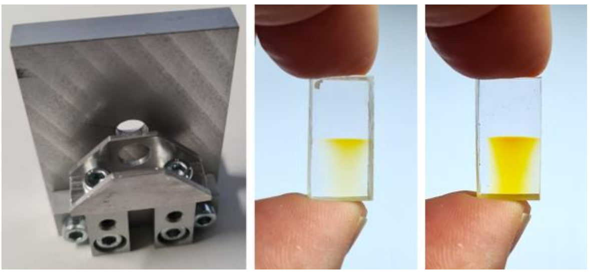

Figure 4. Aluminum collimator with a sample card (left) and MAKROCLEAR dosimeters irradiated diferent doses (right)

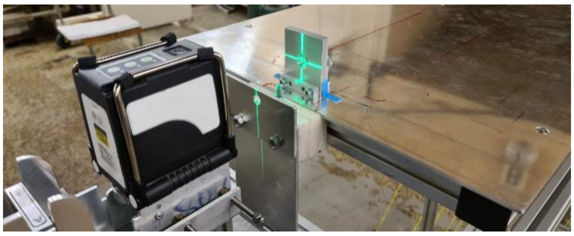

Figure 5. Precise adjustment of the collimator with the sample card to the central axis of the beam, using laser sights

From there, the dose rate at the Bragg maximum at the sample location was determined.

The overall uncertainty can be quantified primarily based on the uncertainties of the gauges used.

The uncertainty of measurements of proton depth dose curves using the Farmer ionization chamber connected to the UNIDOS electrometer from ptw Freiburg depends on several factors:

1. Uncertainty of the ionization chamber (Farmer chamber): The typical measurement uncertainty is around 1% (for common applications). This includes calibration in primary standard laboratory and long-term stability of the chamber.

2. Uncertainty of the electrometer (UNIDOS): The UNIDOS electrometer is a highly accurate instrument with a typical uncertainty of less than 0.5%

3. Calibration constant uncertainty: Calibration constants can generally contribute an uncertainty of about 1% depending on how the chamber was calibrated (eg when calibrating for proton beams). However, for low-energy protons it can reach up to 2%, as the calibration factors in this region may not be as well defined as for higher energies

4. Environmental influence (temperature and pressure, geometric arrangement) may add additional uncertainties of up to 0.5%.

The total combined uncertainty in the dosimetry of proton beams using the Farmer ionization chamber connected to the UNIDOS electrometer is approx. 3% (for a 95% confidence level).

We determined the uncertainty of MAKROCLEAR dosimeters in a reasonable range of doses, e.g. in work [13] to about 10%. Fading of MAKROCLEAR dosimeters was described e.g. in [4], [5], [14]-[16] and can be considered marginal due to the practically immediate evaluation after irradiation.

Individual samples were successively irradiated with 15.5 MeV and 34 MeV protons at doses at Bragg maximum of 0.5 kGy, 2.5 kGy, 5 kGy, 7.5 kGy, 10 kGy.

Another series of samples was irradiated with 17 MeV deuterons at doses at Bragg maximum of 5 kGy, 10 kGy, 15 kGy.

3. Post-irradiation analysis

3.1. Evaluation of irradiated dosimeters using polychromatic light

After irradiation with hadron beams, the dosimeters remain weakly radioactive for several hours (measured by mobile gauge Thermo Scientific FH 40G). The half-life was estimated by measurement to be about 1 hour, so that after 24 hours the activity of the dosimeters was no longer measurable.

A careful cleaning by ethanol and cellulose wadding was followed by scanning of the irradiated dosimeters on an Epson Perfection 850-Pro transmission scanner in white (polychromatic) light with a mean wavelength of 550 nm. The scanning was performed approximately one hour after irradiation, along the beam axis to obtain information about the depth dose curve (Bragg curve) of the respective type of radiation.

3.1.1. Evaluation of dosimeters irradiated with proton beams, in polychromatic light

In the Figure 6 are transmission scans of MACROCLEAR dosimeters irradiated by proton beams with an energy of 34 MeV (left) and 15.5 MeV (right), in doses (from above) 0.5 kGy, 2.5 kGy, 5 kGy, 7.5 kGy, 10 kGy.

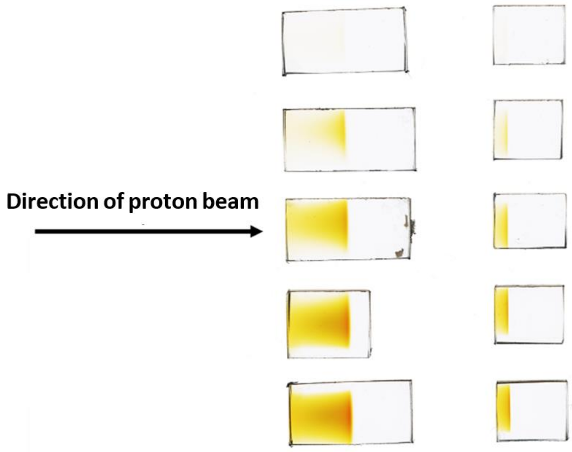

Figure 6. Transmission scans of MAKROCLEAR dosimeters irradiated with a proton beam (coming from the left) with an energy of 34 MeV (left) and 15.5 MeV (right), in doses (from above) 0.5 kGy, 2.5 kGy, 5 kGy, 7.5 kGy, 10 kGy

The bitmaps obtained by scanning were then converted using a specially crafted code in the Ruby programming language into a matrix of values with 8-bit depth according to the degree of darkening. These matrices were imported into an Excel spreadsheet, where the original grayscale images were reconstructed through conditional cell formatting. In the next step, the data was de-noised by low-pass filter convolution (image integration). The convolution kernel consisted of a 3 x 3 matrix.

By accurately measuring the size of each sample with a micrometer gauge, the size of one cell was determined and thus the correct dimensional range of individual elements in the image. This made it possible to construct graphs of the depth curves of optical density and optical density profiles of the individual bundles used, in a dimensional scale corresponding to reality. (Figure 7).

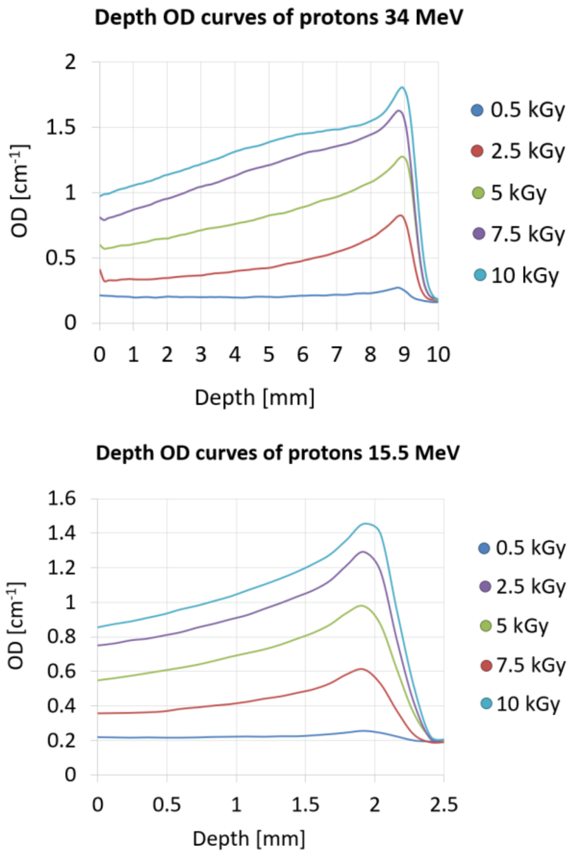

Figure 7. Graphs of optical density (OD) depth curves read from transmission scans of MAKROCLEAR dosimeters - up: depth curves OD of a proton beam with energy 34 MeV, down: depth curves OD of a proton beam with energy 15.5 MeV. The absorbed doses correspond to the individual depth curves, see legend on the right.

A control theoretical model of the proton range and the position of the Bragg peaks was performed in the SRIM calculation program for PMMA (polymethyl methacrylate), which has a slightly lower density (1 180 kg / m3) than PC (1 200 kg / m3) (Figure 8). Despite the excellent agreement between the theoretical model's predictions regarding the proton range and our experimental results, small deviations (not exceeding 0.5 mm) were recorded, which can be attributed to e.g. a few percent difference in the density of both materials.

Optical density (OD - vertical axis in the graphs) was determined as the ratio of absorbance *A*, i.e., the decimal logarithm of the ratio of radiation intensity incident on sample I_0 (mean value of light intensity outside the sample area) and radiation intensity transmitted by sample *I*, and blackening area thickness *d* (so as it was possible to make an absolute comparison of the blackening rate of samples exposed and scanned in different ways):

$$OD = \frac{A}{d} = \frac{1}{d}\log\frac{I_0}{I} \qquad (1)$$

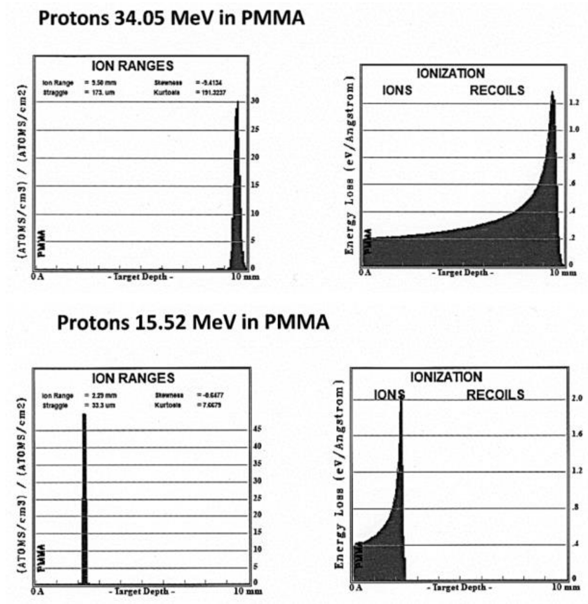

Figure 8: Theoretical model of the range of protons of the mentioned energies in the PMMA material

From Figures 7 and 16, it can be seen that the response of MAKROCLEAR dosimeters in white light is practically linear with a dose in the dose range up to about 7.5 kGy, with a detectability threshold less than 500 Gy. Above this dose, the saturation of the dosimeter is already visible and the optical darkening ceases to be a linear dose, although the various doses are easily optically distinguishable even at values around 10 kGy. This is a range approximately in order smaller than that previously observed with the same dosimeters after exposure to gamma photons of cobalt 60 [4], [5], [13]-[16]. However, this corresponds to the ratio of Linear Energy Transfers (LET) for photons and protons of the energies used.

For radiochromic dosimetry of proton beams, the influence of linear energy transfer (LET) must be considered, because for individual “pristine” peaks, the response of dosimeter in the Bragg peak region can be underestimated by up to 20% (see [7]).

The underestimation is caused by the so-called “quenching” effect, which occurs mainly in the Bragg peak and in the distal part of the extended Bragg peak and is dependent on the linear energy transfer (LET). This effect may be related to the recombination of free radicals in the radiochromic dosimeter [10].

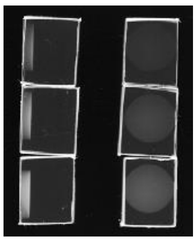

Figure 9. Reconstruction of side (left) and front (right) transmission scans of MAKROCLEAR dosimeters irradiated with a deuteron beam with an energy of 17 MeV with doses in the Bragg maximum (top down) 5 kGy, 10 kGy, 15 kGy

3.1.2. Evaluation of dosimeters irradiated with deuteron beams in polychromatic light

After irradiation with 17 MeV deuterons (Figure 9), the dose response of the dosimeter was slightly weaker than that of the protons, and retained its linearity up to 15 kGy, as shown by the graph. (Figure 10).

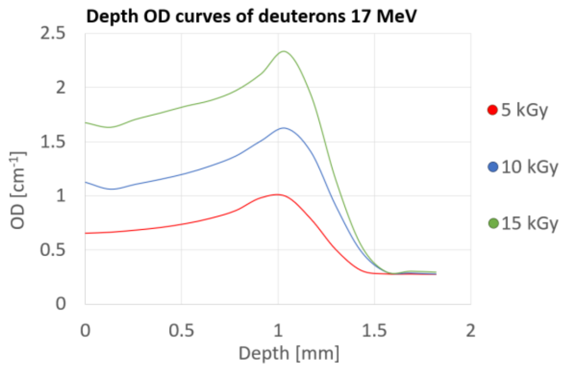

Figure 10. Depth curves of optical density read from MAKROCLEAR dosimeters irradiated with 17 MeV deuteron beams, for three different absorbed doses in the Bragg maximum: 5 kGy, 10 kGy, 15 kGy

The linearity of the dosimeter response to the dose from deuterons, taken at the Bragg peaks, is evident from Figure 11.

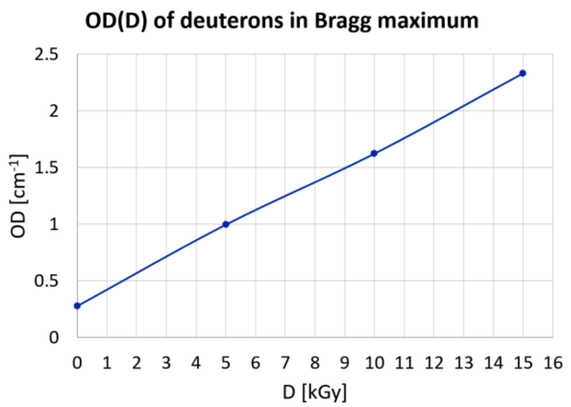

Figure 11. The linearity of the dosimeter response to the dose from deuterons 17 MeV, taken at the Bragg peaks at doses of 5 kGy, 10 kGy and 15 kGy.

The graph (Figure 12) shows the OD profiles of deuterons subtracted in the axes perpendicular to the beam axis - in the x-axis (up) and in the y-axis (down). The graphs indicate a slight misalignment of the collimator outside the central axis of the beam (the central region of the beam with the highest particle density and the highest dose rate), which is about 0.5 mm in the horizontal direction and 1 mm in the vertical direction.

3.2. Evaluation of irradiated dosimeters in monochrome light of different wavelengths

In the next step, approximately two hours after exposure, the irradiated dosimeters were scanned on an Epson Perfection 850-Pro transmission scanner, through a series of color filters transmitting light wavelengths 640 nm, 580 nm, 510 nm, and 450 nm (Figure 13). The scanning was again performed perpendicular to the beam axis to obtain beam dose profile information and along the beam axis to obtain depth dose curve information.

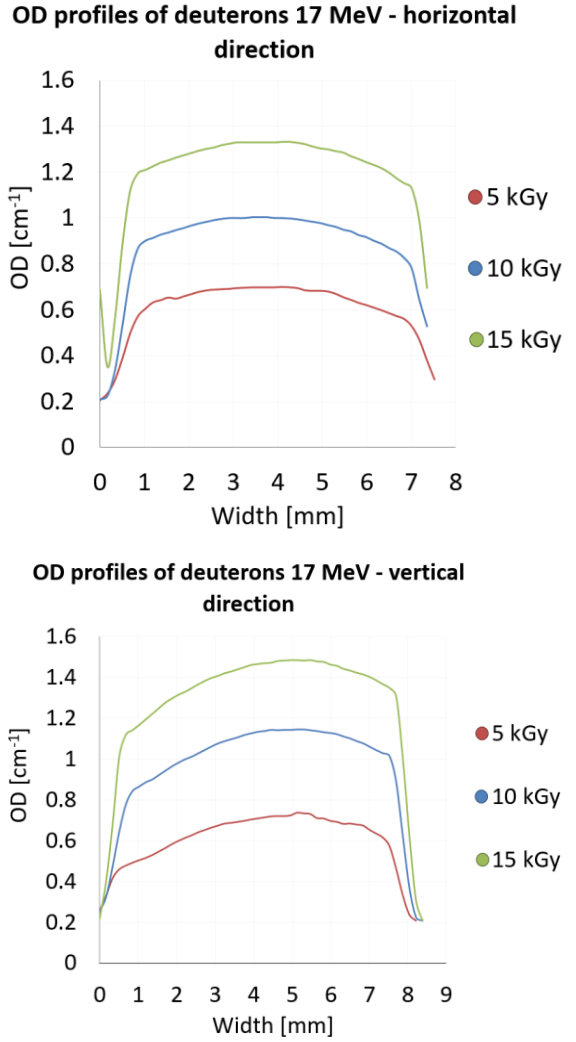

Figure 12: Optical density profiles subtracted from MAKROCLEAR dosimeters for 3 different absorbed doses (see legend on the right) - in the horizontal direction (up) and in the vertical direction (down)

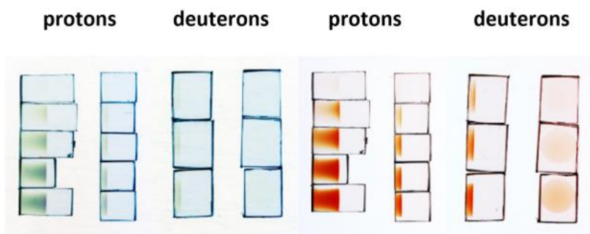

Figure 13. Transmission scans of MAKROCLEAR dosimeters irradiated with protons at doses of 0.5 kGy, 2.5 kGy, 5 kGy, 7.5 kGy, 10 kGy and deuterons at doses of 5 kGy, 10 kGy, 15 kGy, taken at light wavelengths of 640 nm (left) and 450 nm (right) and filtered using the automatic color recovery function, which filtered out the colored background

The acquired images were then adjusted using the automatic color recovery function, which filtered out the colored background (Figure 13). We see in Figure 14 a comparison of the two resulting images taken at wavelengths of 640 nm and 450 nm.

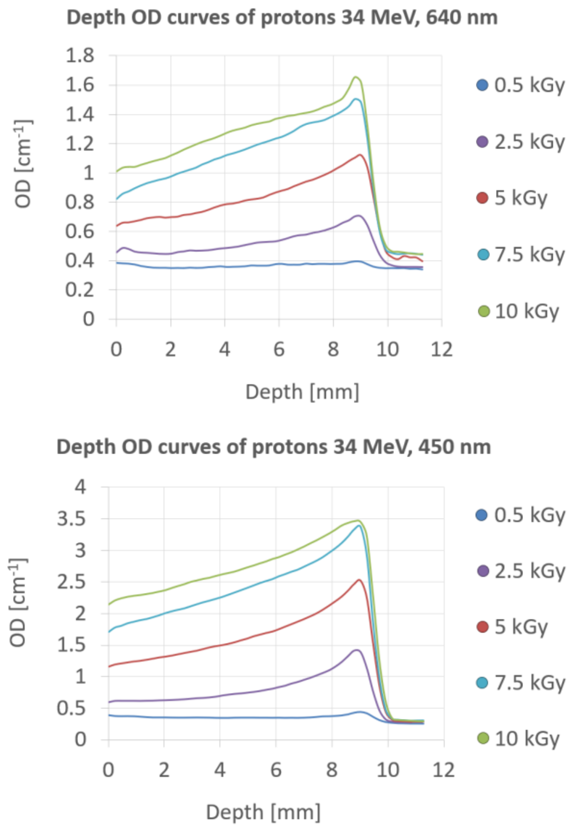

Figure 14: Comparison of depth profiles of optical density of MAKROCLEAR dosimeters irradiated with protons with energy of 34 MeV, read at light wavelength 640 nm (up) and 450 nm (down)

Comparing the Figures 14, it is obvious that the response of the dosimeter is significantly higher at shorter wavelengths of light, regardless of the type and energy of the hadrons used. At the same time, this leads to faster saturation of the dosimeters for shorter wavelengths of light, which is especially evident in higher energy protons. For 34 MeV protons, and 450 nm light, the dosimeter saturates extremely fast already above the 7.5 kGy dose, so that higher doses become practically immeasurable. In contrast, for 640 nm light, the dosimeter response up to 7.5 kGy is practically linear and complete dosimeter saturation occurs much later (Figures 14 and 16). This is a property that have been observed before with MAKROCLEAR dosimeters irradiated by gamma photons or neutrons [1], [4], [5], [13], [16].

For protons with approximately half the energy, the dosimeter response to the dose was lower. Therefore, in 640 nm light, the dosimeter response appeared almost linear up to a dose of 10 kGy, as an optical density of only about 1 cm^{-1} was reached, so it can be assumed that it has not yet approached the saturation point at this dose. In 450 nm light, the deviation from linearity was slightly more pronounced at higher doses, but the dosimeter measured even at a dose of 10 kGy without any problems, because even in this case the point of complete saturation was far from being reached (Figure 15).

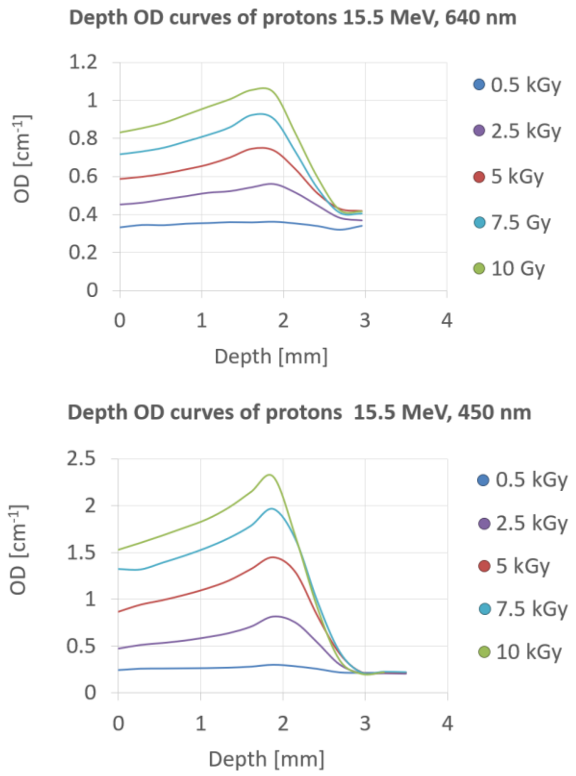

Figure 15: Comparison of the depth curves of the optical density of MAKROCLEAR dosimeters irradiated with 15.5 MeV protons, read at a light wavelength of 640 nm (up) and 450 nm (down)

Figure 16 shows the dosimeter responses to proton doses measured in Bragg maxima. The values were measured in polychromatic light and then in monochromatic light at wavelengths of 450 nm and 640 nm.

It is worth noting that at 640 nm there is a small but measurable change in optical density behind the range of heavy charged particles. This is probably due to other types of particles that have been excited by the hadron beam either in the collimator or in the dosimeter material itself. These can be X-rays, neutrons, and electrons.

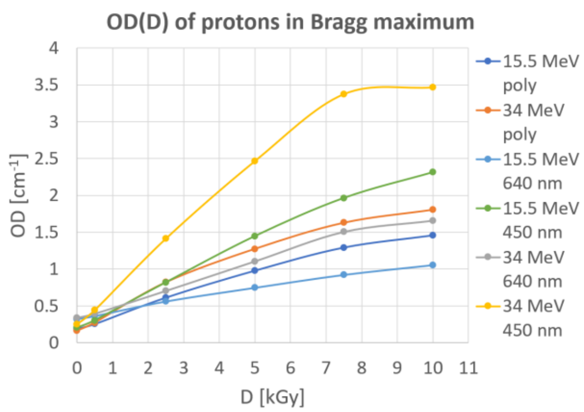

Figure 16. The dosimeter responses to the dose from protons 15.5 MeV and 34 MeV, taken at the Bragg peaks at doses of

0.5 kGy, 2.5 kGy, 5 kGy, 7.5 kGy and 10 kGy. The values obtained in polychromatic light are compared with the values obtained in monochromatic light at wavelengths of 450 nm and 640 nm.

4. CONCLUSION

The results of the analyzes showed that MAKROCLEAR dosimeters are very useful as inexpensive and readily available integrating proton dosimeters in the dose range up to about 7.5 kGy, where their response in white light and monochromatic light with longer wavelenght (about 640 nm) is practically linear with a dose (with a detection threshold less than 500 Gy). An even higher measuring range was recorded in the case of deuterons, where the response of dosimeters is linear with a dose at least to 15 kGy.

The observed OD range is smaller for protons than for photons. Solid-state dosimeters typically do indeed show saturation of the dose response at high LET values. The maximum dose is always less than the expected dose due to the charge recombination bottleneck. This effect is very often observed in hadron monitoring, especially at the Bragg peak [7].

The undeniable advantages of MAKROCLEAR dosimeters compared to other types include their easy preparation as for in the required shape and size, their easy availability, and a very low purchase price (approximately 1 eurocent per piece). Their evaluation is also cheap, easy, fast and does not require expensive or bulky laboratory equipment. 3D information on the hadron beam dose profile and depth dose curve can be obtained in minutes, including the position of the Bragg peak and the dose in the Bragg peak, the ratio of the dose in the Bragg maximum to the dose in the plateau area, and the maximum particle range.

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