KAUNAS UNIVERSITY OF TECHNOLOGY

BENAS GABRIELIS URBONAVIČIUS

DEVELOPMENT AND CHARACTERIZATION OF PLASMONIC SENSORS WITH POLYMER GELS FOR RADIATION DOSE MEASUREMENTS

Summary of Doctoral Dissertation
Technological Sciences, Materials Engineering (T 008)

2019, Kaunas

This doctoral dissertation was prepared at Kaunas University of Technology, Faculty of Mathematics and Natural Sciences, Department of Physics during the period of 2014–2018. Scientific research was partly done in the Institute of Metrology, Institute of Material Science @ KTU as well as Lithuanian Energy Institute and LUHS Oncology hospital.

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KAUNO TECHNOLOGIJOS UNIVERSITETAS

BENAS GABRIELIS URBONAVIČIUS

APŠVITOS DOZĖMS REGISTRUOTI SKIRTŲ PLAZMONINIŲ JUTIKLIŲ SU POLIMERINIAIS GELIAIS KŪRIMAS IR CHARAKTERIZAVIMAS

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1. INTRODUCTION

The essence of modern radiotherapy is the processes of energy transfer and absorption in biological tissue, which determine the use of ionizing radiation for the diagnosis of diseases and the treatment of oncological diseases. Given the fact that radiation doses are always associated with radiobiological effects, even the lowest doses of radiation must be evaluated. Theoretical modeling is most often used for theoretical assessment of interaction processes and planning of treatment procedures, but experimental evaluation is required for the control of effects, which is performed with the help of dosimeters. Dosimeters are made of radiation sensitive materials whose physical and chemical properties change due to exposure to radiation. Among the wide variety of dosimeters (ionizing chambers, films / self-contained films, optically stimulated (OSL), thermoluminescent (TLD), chemical, semiconductor dosimeters) [1], dosimetric gels have exceptional properties because they allow for measurements in a wide range of energies (from tens of keVs) up to tens of MeV) accurately and with good spatial resolution, as well as the ability to capture a three-dimensional dose distribution in the irradiated volume [2, 3]. Dosimetric gels consists of a gelatin matrix and one or more monomers whose degree of polymerisation upon exposure to ionizing radiation is dependent on the absorbed dose. Radiation-induced structural changes in dosimetric gels are associated with changes in the chemical and physical characteristics of the gels which can be measured using nuclear magnetic resonance imaging (MRI), X-ray and optical computed tomography (CTT), optical scanning, sonography and other techniques. Although the NMR imaging method for dosages in irradiated dosimetric gels is considered a "gold standard" for high energy (MeV) radiation therapy, it is complex, costly, and not available in every hospital due to high patient flows.

It should be noted that in case of surface tumors / skin cancer, smaller energy (up to several hundred keV) particles or photons are used, therefore it is especially important to register local surface doses. Usually, for this purpose, "point" TLD or OSL dosimeters and dose readers are used, but they are relatively expensive, with specific limitations for the registration of > 1.0 Gy doses. In addition, when the dosimeter is placed on the patient's skin, the dosimeter also records the particles backscatterd from the patient. Such dosimeters could be successfully replaced by sensors based on surface plasmon resonance (PPR) phenomena, formed from radiation sensitive polymer gels on the surface of the diffractive grating.

The principle of plasmonic sensors is based on the excitation of strong free electron oscillations in the boundary layer between metal and dielectric using polarized light. In the selected characteristic angle or wavelength of the light beam, changes in the intensity of light reflected from the metal-dielectric boundary allows to characterize the chemical and physical properties of the dielectric layer [4, 5]. Plasmonic sensors are commonly used in biotechnology, where the concentration and reaction rate of various organic compounds is assessed [6-8]. No examples of dosimetric sensors based on PPR phenomenon have been found in the literature.

Taking into account the characteristics of known plasmonic sensors, the equivalence of dosimetric gels to biological tissue, and the prospects of creating specific high sensitivity dosimetric gels and the possibility of realizing a direct surface dose registration (without the backscattering component), the concept of gel PPR dosimeter [A1] was proposed and implemented in this work.

This thesis presents the formation of small energy (<250 keV) X-ray photon-sensitive dosimetric gels of acrylamide (AAm) and N,N-methylene-*bis*-acrylamide (BIS) and vinylpyrrolidone and their application in the construction of plasmonic dosimeters for small dose (<10 Gy) registration. Diffraction grating method was chosen to form dosimetric sensors and the original dosimetry system was developed for the dose estimation using a plasmonic dosimeter.

Main aim of this thesis

The aim of the dissertation is to create and characterize plasmonic sensors with polymer gels and adapt them for the registration of small doses of ionizing radiation.

To achieve this goal, the following tasks have been identified:

1. To investigate changes of optical properties of small energy (<250 keV) photon-initiated polymerization processes in dosimetric gels.
2. To evaluate the possibilities of surface plasmon resonance excitation in diffractive structures of optical information media and to functionalize them with dosimetric gels.
3. To create a model of plasmonic sensor scanning system and apply it to the evaluation of medical exposure doses.
4. Verify dosimetric measurements using plasmonic sensors.

Originality of this work

1. Modified nPAG and VIPET dosimetric gels containing N,N-*bis*-acrylamide have been shown to be sensitive to low energy (<250 keV) photon exposure and that their sensitivity in low dose range (0.5 to 5 Gy) increases by 11% compared to baseline (selected based on literature analysis) gels.
2. It has been shown that modified gels can be used to functionalize the surface of optical information media used as a structural element of a plasmonic sensor.
3. A plasmonic sensor readout system was developed for dosimetry measurements, allowing measurements to be made in real time.
4. For the first time, it has been demonstrated that using a functionalized plasmonic surface with dosimetric gels, a point dosimeter is implemented

that allows for direct estimation of the surface dose (without the backscattering component) and having stable metrological characteristics of in small energy (keV) and small dose (<5Gy) range.

Authors' part

1. Production of experimental gels and experimental characterization of their properties.
2. Formation of plasmonic dosimetric sensors on the diffraction grating of the optical information medium.
3. Development and metrological evaluation of plasmonic dosimetric sensor readout system.
5. Creation of dose estimation method using plasmonic sensor.

Photoscanning measurements were conducted by MB „Šeši partneriai“ (Dr. Nergina Šeperienė)
Raman spectrometric measurements were conducted at the Institute of Materials Science, Kaunas University of Technology (Dr. Asta Tamulevičienė)
Measurements with scanning electronic microscope were conducted at Lithuanian Energy Institute (Dr. Mantas Sriubas)

Structure of the thesis

Dissertation consists of introduction, literature review, research methodology, results discussion sections, conclusions, references. Volume of the dissertation is 114 pages. It contains 74 figures, 13 tables and 215 references.

Dissertation approbation

The results of the research on the topic of the dissertation were published in 7 scientific publications: 4 articles were published in publications containing the citation index in the Web of Science Clarivate Analytics database; 4 articles were published in conference publications with CPCI status in the Clarivate Analytics database. The results of the work were presented at 7 international conferences.
During the preparation of the dissertation, 2 articles on metrology were additionally published in the journals included in the Web of Science Clarivate Analytics database.

1.1. History of Surface Plasmon Resonance

Surface Plasmon Resonance (SPR) is widely used in chemical and bio-sensors. The SPR-based sensor itself was first developed around 80 years ago, when the SPR phenomenon was first detected [1]. First applied SPR research was directed towards gas detection sensors [2]. This achievement was quite significant in comparison with other sensor systems, since it was characterized by high sensitivity and the ability to measure parameters in real time. Fundamental principle of SPR sensors is based on strong electromagnetic field oscillations in the metal-dielectric boundary when the perpendicularly polarized light hits this plane. This strong oscillation manifests itself in how the reflected light intensity drops at a certain angle of incidence or wavelength. Most innovations in this field have occurred over the last three decades, when plasmonic sensors have been practically applied in the fields of chemistry, food, environmental monitoring and medicine [4-7].

1.1.2. Plasmonic Sensor Readout Methods and Parameters

When reading out plasmonic sensors, a sensogram is obtained which can be analyzed in real time, or after complete readout of the sensor. This is done regardless of the readout method. There are 4 main methods for extracting PPR signal.

The first method is based on intensity modulation. This method is based on measuring the intensity of light reflected from the sensor. Intensity can be related to the angle of incidence to the sensor, or, at a fixed angle, associated with the change in wavelength. A sensogram of this method is shown in Figure 1. Visual information is captured by detecting light intensity in a detector matrix (eg CCD). It can be presented even as a video or a photo.

Second SPR excitation method is based on angular modulation. This method is based on the change of intensity of monochromatic light, reflected from the plasmonic sensor at different angles. Position of the minima of the intensity (in the angular space) depends on the refractive index of the material on the surface of the sensor.

Third method is based on wavelength modulation. This approach is based on spectral measurements of light reflected from the sensor. When the light is reflected from the plasmonic sensor, a certain wavelength will be absorbed (in the case of a fixed geometry), which will be clearly visible in the received spectrum.

The fourth method is based on phase modulation (Fig. 1b). This method uses a monochromatic light source and phase shift measurement equipment, for example. phase amplifier. In addition to this additional equipment, the optical system of this readout method is much more complex compared to the ones discussed prior. These technical difficulties are reflected in a relatively small number of studies using such a readout system, and there are no such commercialized systems in general.

Typical sensograms for all of these sensor readout methods are shown in Figures 1 and 2.

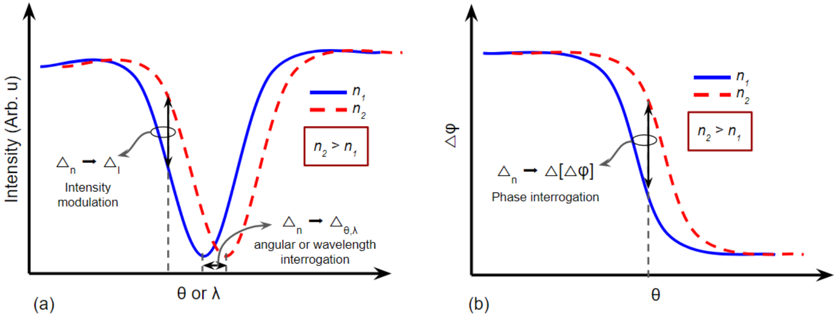


Figure 1. a) typical sensory intensity, angular and wavelength modulation, b) A typical sensor for phase modulation

One of the advantages of plasmonic sensors is the ability to measure changes in the refractive index of the sensitive layer (dielectric) in real time. Real-time measurements sensograms are shown in Figure 2.

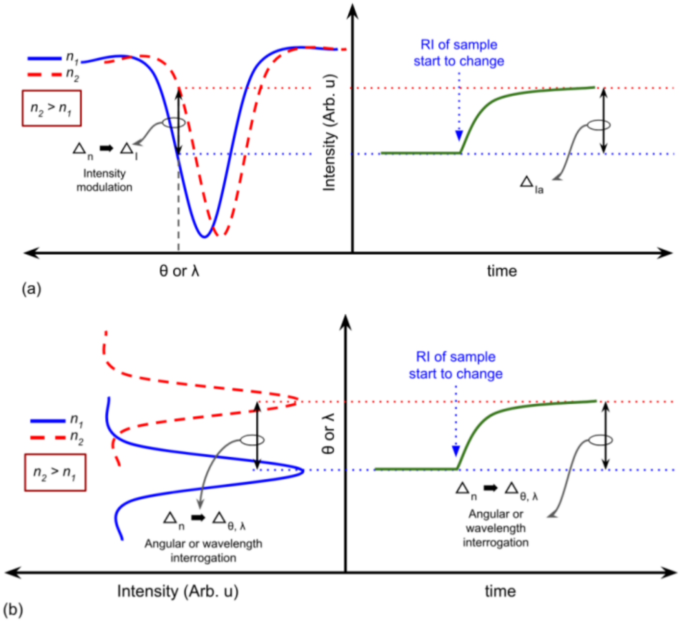


Figure 2. Typical sensors for measuring refractive index changes in real time a) Intensity modulation case b) Waveform or angular modulation case

Optical systems for plasmonic sensors

Several optical systems are available for SPR excitation. The response of the plasmonic sensor corresponds to the change in the characteristics of the light reflected from the sensor, as discussed in the previous section. Depending on the technical solution, SPR sensors can be grouped based on the optical excitation method:

- Prism coupling
- Diffraction grating coupling
- Waveguide coupling

Schematically, these solutions are depicted in Fig. 3

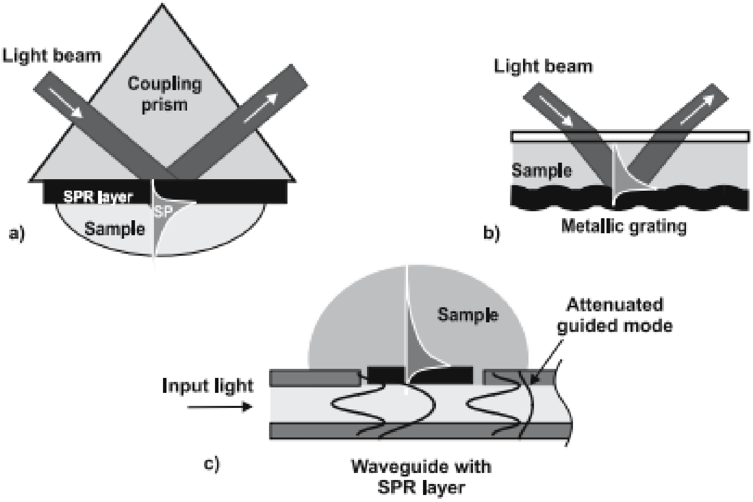


Figure 3. Plasmonic sensors with a) excitation with a prism, b) excitation with a diffraction grating, c) excitation fiber [8]

1.2. Dosimetric Gels

One of the goals of radiotherapy is to homogenously “cover” the tumor with a suitable absorbed dose, while minimizing the dose to surrounding healthy tissues [9,10]. Technologies currently used in radiation oncology make it possible to ensure proper distribution of doses in malignant tissues [11].

It is important to remember that any inconsistency in this case would lead to lower irradiation of the cancerous tissue and potentially higher doses to vital organs located next to it. For this reason, the possibility of checking the distribution of doses in a three-dimensional space, in an radiologically equivalent material to biological tissue, before the radiotherapy procedure, can reduce the likelihood of such errors. [12]

To assess the complicated distribution of doses seen in modern radiotherapy techniques, a dosimetry method is necessary that allows for accurate measurement of doses in three dimensional space and, of course, with good spatial resolution. In clinical radiotherapy, gel dosimeters have a high resolution and precise 3D dose screening, and allows to compare the measurement results with the data provided by the dose planning system. Meanwhile, most current dosimetric methods allow dose measurements to be made only in one-dimensional (point-to-point) or two-dimensional space (ex. thermoluminescent dosimeters, ionization chambers and radiographic films) [13]. Dosimetric gels consist of several different chemical compounds that are sensitive to ionizing radiation. When irradiated, the polymerization degree of dosimetric gels changes, and is proportional to the absorbed dose [14]. Ionizing radiation in dosimetric gels causes structural changes, which also change certain gel properties, such as the duration of proton relaxation (during magnetic resonance scanning), physical density, optical density, elasticity [15]. These changes can be measured using magnetic resonance scanner [16], computed tomography [17], optical scanning [18], ultrasound methods [19]. Gel dosimeters have useful properties that can simplify radiotherapy dosimetry, especially in conditions where conventional dosimetric methods are lacking [20]. These features are: the ability to measure complex three-dimensional dose distributions; Equivalence to biological tissue in the radiation field; response independence to the angle of exposure, high spatial resolution, dose integration during procedures, etc. [21,22]. Dosimetric gels are relatively safe and easy to make and use, although it also contains toxic substances such as acrylamide, which must be used with appropriate protective equipment.

Application of gels involves the usual dosimetry - distribution of the doses, determination of doses next to the irradiation field; distribution of doses in various diagnostic procedures, distribution of doses in various types of radiotherapy: in classical radiotherapy, in intensity modulated radiotherapy and in stereotactic radiosurgery; distribution of doses around various brachytherapy sources, and the evaluation of inhomogeneity in biological tissue.

As discussed previously, combining dosimetric gel advantages with plasmonic sensor readout sensitivity would make a new type of dosimetric sensor that will possibly have highly advantageous properties for medical applications.

2. FORMATION OF PLASMONIC DOSIMETRIC SENSORS

Literature analysis allowed to propose the concept of simple, cost-effective, plasmonic sensor that could be used as a dosimeter [A1]. Based on this concept, the plasmonic sensor is formed on the diffractive structure of the commercial optical media by coating the surface with a dosimetric hydrogel, which undergoes polymerization under ionizing radiation. The degree of polymerization of the hydrogel depends on the type of ionizing radiation and the parameters of exposure and on the energy absorbed by the irradiated gel.

The process of forming and functionalizing sensors can be divided into three main stages:

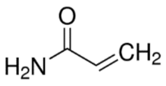
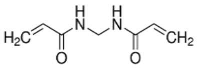
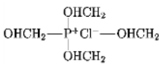
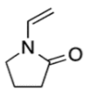
1. Production and irradiation of dosimetric gels;
2. Functionalization of the commercial optical media diffractive structure for forming plasmonic dosimetric sensors;
3. Formation of plasmonic sensors.

2.1. Preparation of Dosimetric Gels

Based on the literature review [17,23-26], main components of the dosimetric gels were selected, which are the monomers localized in the gelatin matrix, which tend to polymerize under the influence of ionizing radiation, monomers promoting crosslinking, oxygen scavenger and other special additives. These gels exhibit pronounced spatial dosimetric characteristics, because under the ionizing radiation gel polymerization takes place, the degree and other characteristics of which depend on the energy absorbed in the irradiated volume. NPAG and VIPET polymer gels were used to create plasmonic dosimetric sensors. This choice was determined by the level of research on the main properties of these types of gels, discussed in the literature review, and experimental experience [27-29].

Chemical composition of nPAG and VIPET polymer gels used to form experimental samples is presented in Table 1.

Table 1. Chemical composition of dosimetric gels

	Water	Gelatin	Monomer	Crosslinker	Oxygen scavenger
nPAG	Distil. Water (≥99% HPLC, (Sigma Aldrich)	Porcine (A type, 300 bloom (Sigma-Aldrich)	Acrylamide, (AAm), ≥99% powder, Sigma Alrich 	*N*,*N*- metilen-*bis*-acrylamide (BIS), 99% powder, Sigma-Aldrich 	Tetrakis-hidroksimetil phosphor chloride (THPC), 80% H_2O solution (Sigma-Aldrich)  $OHCH_2-\overset{OHCH_2}{\underset{OHCH_2}{P^+}}Cl^- -OHCH_2$
VIPET			*N*-vinil-pirolidon (VIPE), ≥99% Sigma –Aldrich 		

2.1.1. Gel Irradiation with Small Energy X-Ray Photons

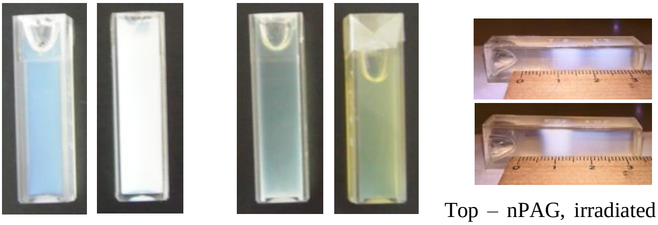
Dosimetric gels were irradiated at the Lithuanian University of Health Sciences Hospital, Kaunas Clinics, Oncology Hospital, using the GULMAY D3225 X-ray device (Fig. 4).

a) b)

Figure 4. a) short-range X-ray therapy device GULMAY D3325, b) GULMAY D3325 control panel with monitoring equipment

When assessing the effects of low energy and low dose X-ray radiation on the polymerization of dosimetric gels, samples were irradiated by selecting an X-ray generator voltage from 150-250 keV and maintaining a constant current of 20 mA. To reduce the impact of very low energy photons, a 2.33 mm Cu filter was used. During the irradiation, a rectangular applicator was used to form (10cm x 10cm) exposure field in which samples were placed. Such exposure parameters were selected to provide a homogeneous exposure to the entire gel volume in a sample cuvette.

Experimental samples were irradiated with doses 0-5 Gy, because information on low energy X-ray polymerization processes in gels, when irradiated with low doses, is sparse. Absorbed dose was controlled using a calibrated ionizing chamber Farmer 30013 (PTW Freiburg) which was placed in the center of the exposure field together with samples. Some results of such irradiation is shown in Fig. 5.



nPAG, 0 Gy

nPAG, 5,0 Gy

VIPET, 0 Gy

VIPET, 5,0 Gy

Top – nPAG, irradiated with 1.5 Gy and 2 Gy
Bottom – nPAG, irradiated with 3 Gy

Figure 5. Examples of nPAG and VIPET gels before exposure and after exposure to low energy X-ray

2.2. Determination of The Suitability of The Diffraction Gratings Used in Optical Media for Plasmonic Sensors

Effectiveness of the diffraction grating of optical media, for SPR excitation, can be easily verified by applying the models discussed in the literature review of this dissertation, because the geometric parameters of known optical storage media (Fig. 6) satisfy the surface plasmon resonance condition: $h/\Lambda < 0.15$.

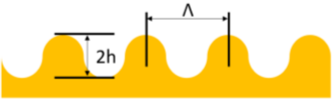


Figure 6. Geometrical parameters of a diffraction grating

Condition $h/\Lambda < 0.15$ is significant only when examining surface plasmon resonance imaging on a non-functionalized diffraction grating. Functionalization of the grating (after the formation of the sensitive surface layer) also changes the conditions of excitation of surface plasmon resonance. Since the response of the formed dosimetric sensor is ultimately evaluated by the change of the refractive index of the sensitive layer, it is important to take into account the measurement characteristics of this measurand.

Absolute value (or its change) of the refractive index in the plasmonic sensors can be estimated from the following diagram (Figure 7).

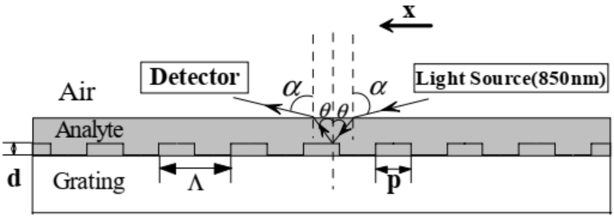


Figure 7. Schematic of Dosimetric Gel Refractive Index Change [30]

Λ is the period of the metallic grating, p is the width of the grating, d is the depth of the grating. Sometimes the parameter f in the calculations is called duty cycle and is expressed as f = p / Λ. Assuming that the grating does not change the excitation wave propagation characteristics (as discussed in the literature review), it is possible to write and equation the described the law of impulse conservation in the system [30]:

$$n_a \sin\theta_R + m\frac{\lambda}{\Lambda} = \pm\sqrt{\frac{\varepsilon_m n_a^2}{\varepsilon_m + n_a^2}} \tag{1}$$

Here θ_R is the resonant fall angle, ε_m is the relative permeability of the diffraction grating metal, n_a is the refractive index of the dielectric on the grating (sensitive layer). m may be positive or negative depending on which diffraction order is considered.

Knowing the wavelength of the excitation light used and the maximum expected refractive index value, the suitability of commercial optical media for forming plasmonic sensors was modelled.

Based on this calculated data and formula 1, it is possible to model the range of incidence angles where the expected response of the dosimetric sensors is observed. Results of this modeling are presented in Figure 8.

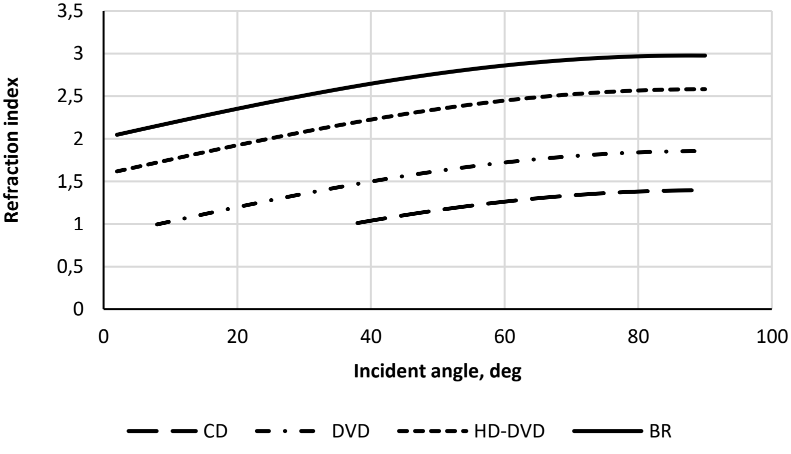


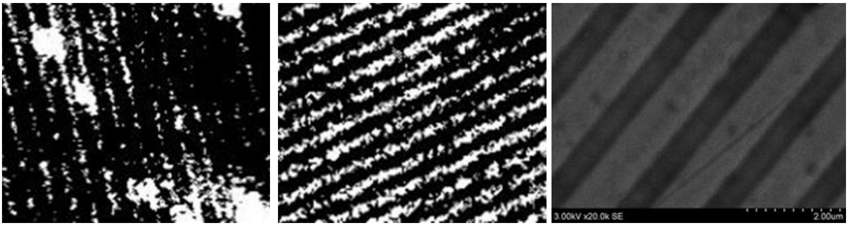
Figure 8. Angle range for different optical media in which refractive index changes can be measured

For the simulation, a range of 10^o to 90^o incidence angles was selected. The graph in Figure 8 shows that DVD grating has the largest range of useful angles, while the CD grating is only suitable for measurements of smaller refractive indexes

(<1.5). Meanwhile, HD-DVD and BlueRay disc gratings are only suitable for larger (> 1.5) refractive index measurements with the selected monochromatic light source.

2.3. Preparation of Optical Media Diffraction Gratings

In order to use the diffraction gratings of an optical disc for the observtion of surface plasmons, it is necessary to remove its protective layer. ABRO PR-600 paint remover was used to remove the protective lacquer layer of off the optical media diffraction gratings. Cleanliness of the extracted diffraction grating was first evaluated with the Opta-Tech optical microscope MN800. Level of surface contamination was determined using the automatic identification method. Figure 9 shows the images of the baseline and “dirty” diffraction gratings after edge detection mask application. For comparison, a SEM image of the cleaned diffraction grating is presented



a b c

Figure 9. The images of the CD diffraction grid with the edge recognition mask are: a) "dirty" image, b) baseline image, c) SEM scan of the grating

It can be seen from the SEM image that the level of the contamination of the grating surface, which was confirmed by the analysis with the optical microscope, meets the requirements.

In order to evaluate the periodicity of the lattice, cleaned surface was examined with a profilometer XP-200 (Fig. 10).

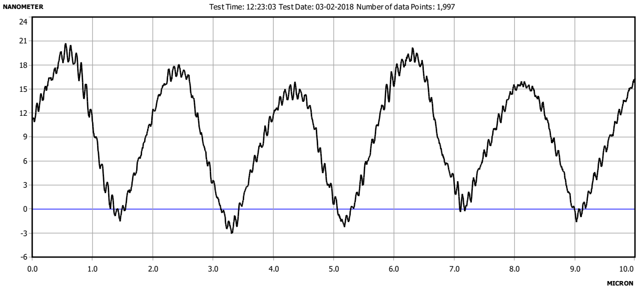
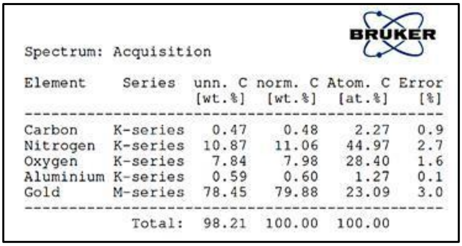


Figure 10. Periodicity study of optical media cell

An additional analysis of the gratings' chemical composition confirmed (Fig. 11) that the predominant component of the grating is gold, which is ~ 80% by weight.



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                                              BRUKER
Spectrum: Acquisition

Element    Series  unn. C norm. C Atom. C Error
                   [wt.%]  [wt.%]  [at.%]   [%]
-----------------------------------------------
Carbon    K-series   0.47    0.48    2.27   0.9
Nitrogen  K-series  10.87   11.06   44.97   2.7
Oxygen    K-series   7.84    7.98   28.40   1.6
Aluminium K-series   0.59    0.60    1.27   0.1
Gold      M-series  78.45   79.88   23.09   3.0
-----------------------------------------------
            Total:  98.21  100.00  100.00
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Figure 11. Elemental composition of the optical media diffraction grating

Cleaned optical disc diffraction gratings were cut into 10x10 mm segments and then used as a basis for plasmonic dosimetric sensors. Cutting was carried out using the hotwire method and using a guillotine.

Samples of the optical disc cut by both methods were evaluated using a microscope MN-800. Measurement were done "over the edge". The images obtained are shown in Figure 12.

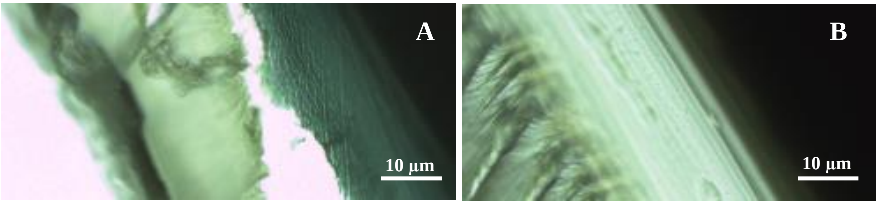


Figure 12. Edge of the cleaned diffractive structure cut by: using the hot wire method (A) and guillotine (B).

Obviously, the application of a hot wire method has failed because of a clear deformation of the boundary between the polymer backing and the diffractive structure.

2.4. Development of a Plasmonic Sensor Readout System Based on Angular Modulation

Angular modulation plasmonic sensor readout system does not require precise mechanical elements (goniometers) to determine the angle of incidence/reflection of the light used. A monochromatic light source can also be used, so the optical system consists of a smaller number of elements. Light registration unit also has other technical requirements that may be more practical in individual cases. Fig. 13 is a schematic view of the system. Main elements of the system are discussed below

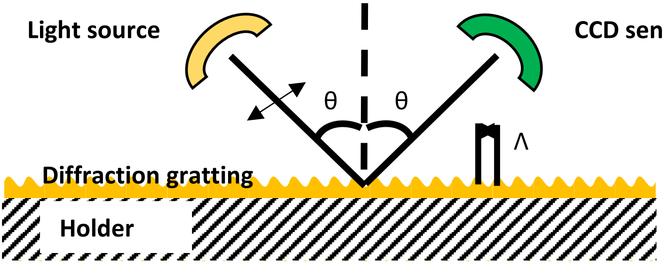


Figure 13. Schematic view of an angular modulation based system

He-Ne laser with a wavelength of 632.8 nm was chosen as a light source. Using a laser as a light source ensures good collimation. Used laser has an integrated polarizer, but an additional polarizer was added to the system for better polarization performance.

Canon CLG Series CCD was chosen to measure light intensity. This device is used as a contact image sensor in various electronic devices. Sensor itself is compact, sensing elements form one row. This arrangement of sensitive points (one row) allows for measurements to be performed at many angles with readjusting the system. Optical resolution - 600 pixels per inch (one pixel effective dimension ~ 42µm). This sensor's optical resolution ensures angular resolution greater than 0.1° in terms of incident light angle. Since the sensor is designed to measure light intensity only in perpendicular direction, it has been modified by removing the optical apertures.

In the initial version of the measurement system, an ARM architecture processor with Samsung embedded software was used to control the CCD. Data was scanned in uncompressed image format (BMP) using special Samsung software for Windows.

Since the operating principle of this system is based on changing the angle of light incidence to the diffractive grating, an angular modulator was used. Switec stepper motor was selected for this purpose. Its' maximum turning angle is 230°. A plate for the test samples was attached to the motor shaft. Design of the measuring system consists of a steel base with a plastic construction for electronics. Neodymium N48 magnets are used for positioning the CCD sensor. Since all electronic system components use an USB interface for data transfer, a USB hub is used to connect all components. The overall system view is shown in Figure 14.

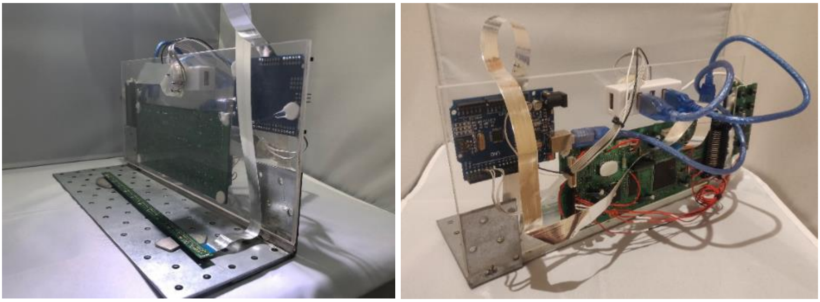


Figure 14. Angular modulation system a) general view, b) rear view

During later experiments this angular modulation plasmonic sensor readout system was updated to increase data processing efficiency. Originally data is received as an image file (.BMP format) and then processed by the MATLAB software package. This last step takes a relatively long time and complicates data processing, especially with a lot of measurement results, as discussed in the following sections. For this reason, the system has been upgraded so that the initial data processing is performed by a newly programmed CCD controller.

After the measurement, the results of the CCD scan are automatically displayed, so no additional processing operations are required. The overall view of the system after the update is presented in Figure 15.

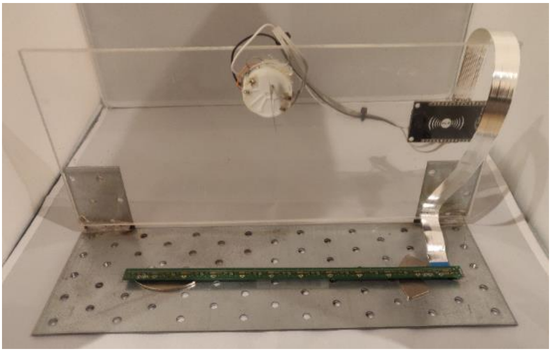


Figure 15. View of an updated angular modulation system

3. SURFACE PLASMON RESONANCE MEASUREMENTS IN A DIFFRACTION GRATING- DOSIMETRIC GEL STRUCTURE

A centrifuge method was used to form the dosimetric gel layer on the prepared substrate with a diffractive structure. This method is simple, with the ability to control the layer setting and good repeatability [31

Because the discussed angular modulation measument system (like all similar measurement systems) has a light sensor with a finite sensitivity. It means that there is thickest layer of dosimetric gel at which changes in the intensity of the light reflected by the plasmonic sensor can still be recorded. To determine this maximum layer thickness, a series of sensors were created using the DVD disc diffraction gratings and nPAG and VIPET dosimetric gels. Based on previous experiments, sensors with a layer of dosimetric gel ranging from 1 to 30 µm were formed. Dosimetric layer thickness step is selected at 1µm. For each layer thickness 3 separate sensors were formed, for repeated measurements.

Due to self-polymerization (because of oxygen trapped in the gel during its making) measurements were made after about 24 hours after the formation of the sensors. Figure 16 and 17 show the readout results from the sensors with VIPET and nPAG dosimetric gels.

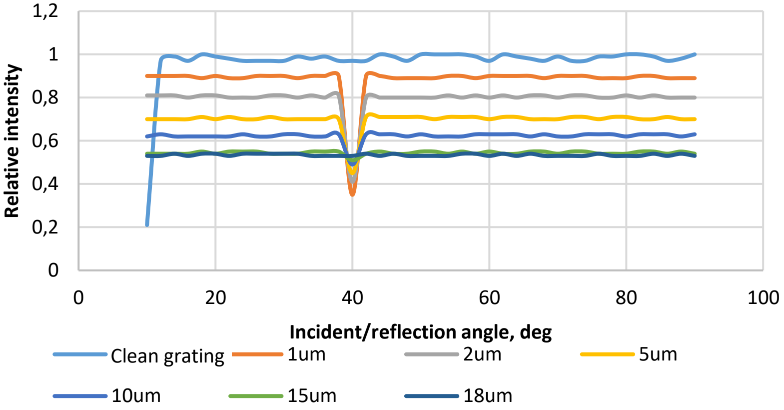


Figure 16. Readout results of plasmonic sensors with different thicknesses of the VIPET dosimetric gel layer

Graphically, the results of the clean diffraction grating are also shown. It can be seen that the SPR of clean DVD disc grating is visible in the area of the ~ 10^o incident angle, which coincides well with the results of the previous mathematical calculation. Effect of different thickness of the dosimetric layer on the measurement results is also given below. Readout system is not sensitive enough to measure the change in light intensity, when dosimetric layer thickness is higher than 18µm. It is seen that with VIPET dosimetric gel SPR is observed at an incident angle of ~40^o, which corresponds to a refractive index of 1.497. Theoretical refractive index of vinylpyrrolidone (the highest refractive index for VIPET dosimetric gel) is 1.501 [19].

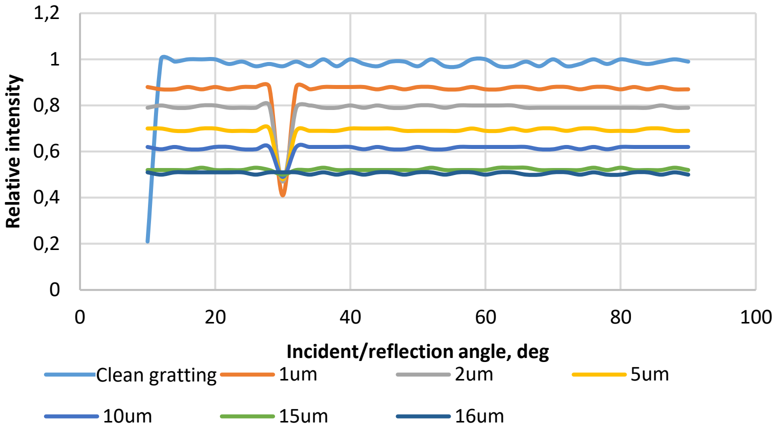


Figure 17 Readout results of plasmonic sensors with different thicknesses of the nPAG dosimetric gel layer

In the case of nPAG dosimetric gel, SPR is observed at an incident angle of 30°, which corresponds to a refractive index of 1.365. Theoretical refractive index of acrylamide (the highest refractive index substance in nPAG dosimetric gel) is 1.38. It appears that these initial results indicate a good coincidence between experimental and theoretically expected measurement results. When analyzing the effect of layer thickness on measurement results, it is evident that as the thickness of the layer increases, the absorption of the incident light is increased, which is reflected by the decrease of the recorded (reflected from the sample) light intensity. At the layer thickness value of the 18µm (VIPET) and 16µm (nPAG) it is virtually impossible to distinguish the area in which SPR occurs. For this reason, results of a higher layer thickness are not provided.

3.1. Influence of Low Energy X-Ray Photons on the Properties of Dosimetric Gels

According to Buger-Bero-Lambert's law, light absorption depends on the density of the medium to be analyzed, the concentration of various molecular-functional groups and the thickness of the sample. Considering that radiation-induced polymerization in the dosimetric gel is associated with new structural fragments that influence changes in gel optical density, it is possible to indirectly determine the degree of gel polymerization from UV-VIS absorption spectra. For this purpose, UV-VIS absorption spectra of nPAG and VIPET gels irradiated with different doses in the GULMAY D3225X device were analyzed. Characteristic examples of nPAG and VIPET spectra are shown in Figure 18 and 19 respectively. It should be emphasized that the effect of the cuvette in the spectra was eliminated automatically.

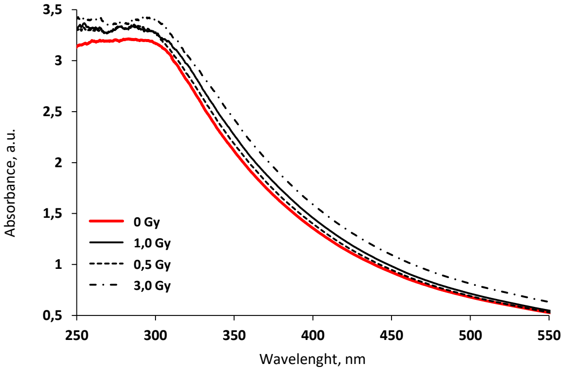


Figure 18. Absorption spectrum of nPAG samples irradiated with 220 keV energy X-ray photons

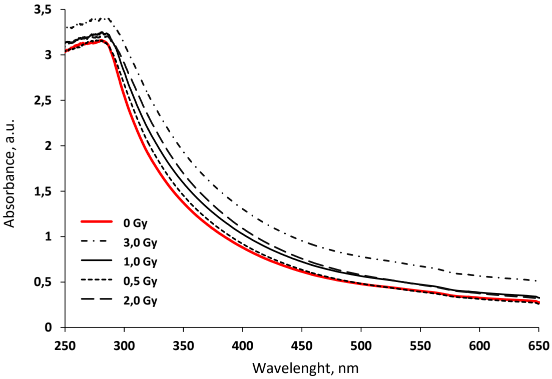


Figure 19. Absorption spectrum of VIPET samples irradiated with 220 keV energy X-ray photons

This study showed that using the optical spectroscopy method, 0.0997 Gy^{-1} dosimetric sensitivity for nPAG gel and 0.1804 Gy^{-1} dosimetric sensitivity for VIPET gel were detected (Figure 21a). These sensitivity values correlate well with the work of other authors [18,19], as well as with the sensitivity values determined using the dose photodetector [30]: 0.0949 Gy^{-1} and 0.1762 Gy^{-1} for VIPET gel and for nPAG gel respectively. Based on the results of this study, it can be assumed that nPAG and VIPET gels are sufficiently sensitive to low-energy X-rays.

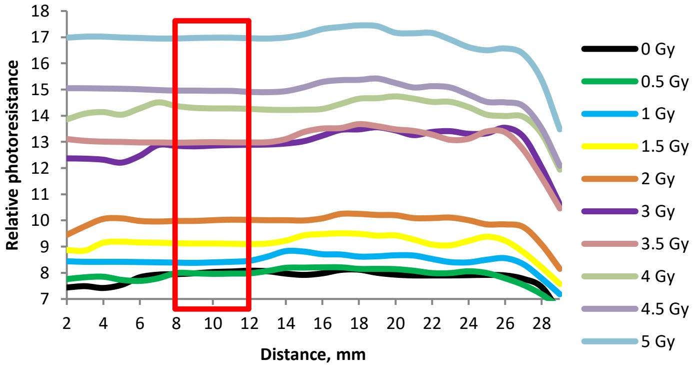


Figure 20 Dose profiles of nPAG gel irradiated with different doses as scanned along the cuvette

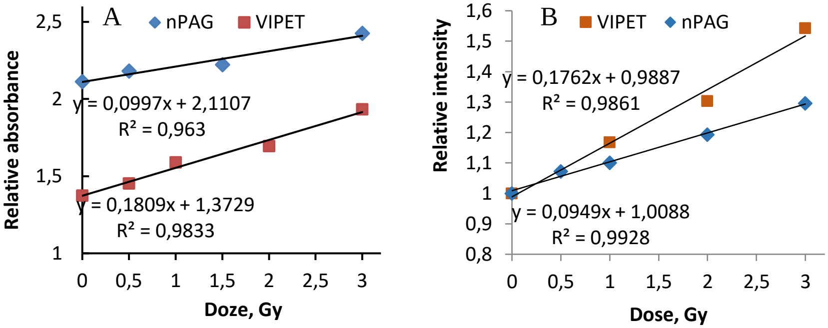
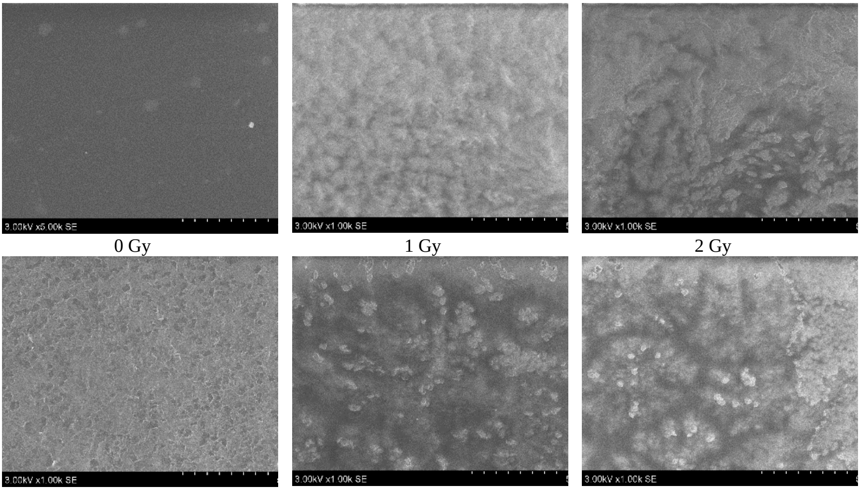


Figure 21 Dependence of dosimetric sensitivity of nPAG and VIPET on absorbed dose: A - with UV-VIS spectroscopy; B - with photo scanning method [27]

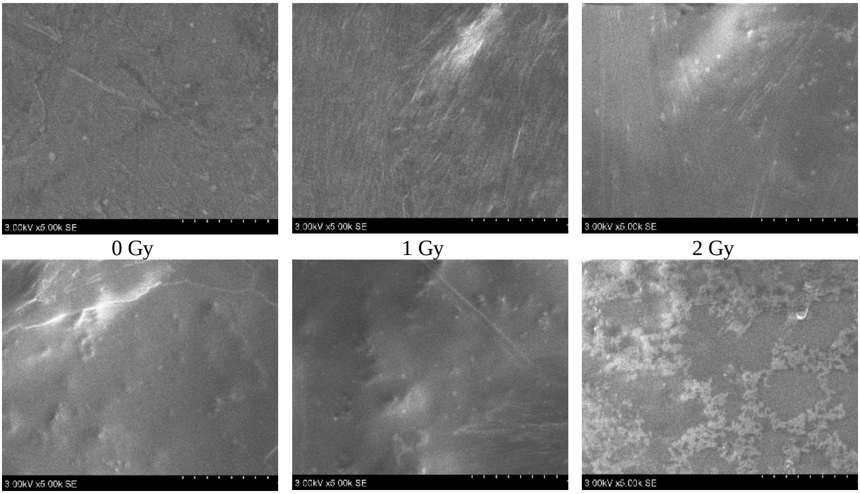
In addition, the results of optical measurements can indirectly address structural changes in the irradiated gels due to polymerization. It is known that the intensity and position of the spectral absorption peaks depend on the chemical composition of the gel and its molecular structure [31]. In X-ray irradiated samples, it can be seen that the absorption increases with the increase of the irradiation dose, and the absorption curves spreads slightly (Fig. 18 and Fig. 19). These tendencies can be associated with gel polymerization [31]. Additionally a set of SEM images made (Fig. 22 and Fig. 23) that shows that even with the relatively small thickness of the dosimetric gel layer (5µm), structural changes seen on the surface tend to be related to the absorbed dose. These images reveal changes on the surface of nPAG and VIPET gels, which are likely to be related to the molecular structure of monomers. These results correlate with the work of other authors [13, 19], which deal with low dose (<10 Gy) irradiated nPAG gels with less than 10% BIS concentration. It is claimed that this gel forms a grainy structure that is associated with gel surface changes [19].



0 Gy 1 Gy 2 Gy

3 Gy 4 Gy 5 Gy

Figure 22 SEM images of nPAG gels irradiated at different doses



0 Gy 1 Gy 2 Gy

3 Gy 4 Gy 5 Gy

Figure 23 SEM images of VIPET gels irradiated at different doses

3.2. Application of Surface Plasmon Resonance for Characterization of Irradiated Gels

In order to form plasmonic sensors for use in medical diagnostics and/or therapy, it is necessary to carry out an investigation of their properties as a dosimetry method. For this purpose, a series of sensors were formed, with dosimetric gel layer thicknesses selected to be 1, 2, 5, 10 and 15 Prepared dosimeters were irradiated with 120 keV photon radiation at the X-ray therapy device GULMAY D3225 with doses [0.5; 10] Gy at a dose rate of 2 Gy/min. For each layer thickness, 60 sensors were formed to provide three sensors for each absorbed dose. All sensors (dosimeters) were readout before irradiation and evaluated for their response - SPR excitation incidence angle. For the residual polymerization processes in the dosimetric gel, all dosimeters were readout 24 hours after irradiation. Change in the relative intensity of the response signal of plasmonic dosimeters with VIPET gel irradiated at different doses is shown in Figure 24 and with nPAG gel in Figure 25. Refraction index vs absorbed dose for nPAG and VIPET gels are shown in figures 26 and 27.

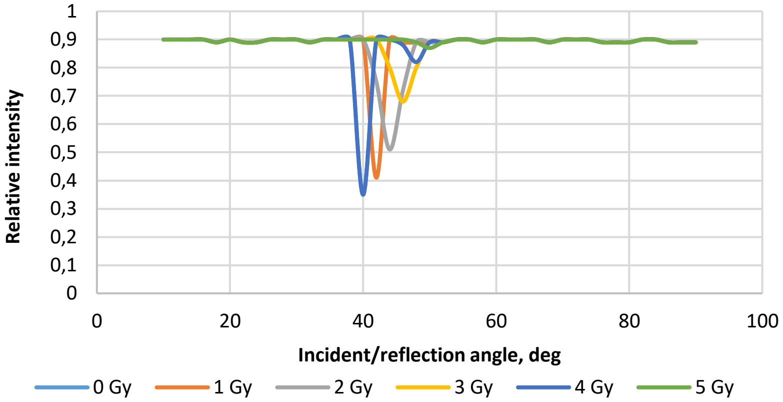


Figure 24 Plasmonic dosimeter, with VIPET dosimetric gel, response at different absorbed doses

Figure 25 Plasmonic dosimeter, with nPAG dosimetric gel, response at different absorbed doses

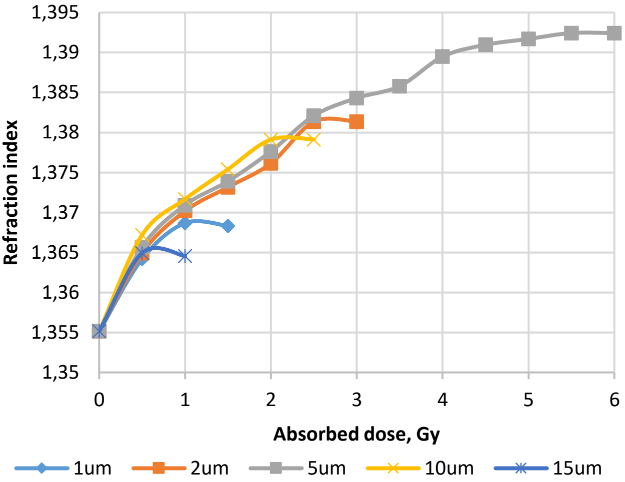


Figure 26. Dependence of refractive index on absorbed dose of nPAG gel

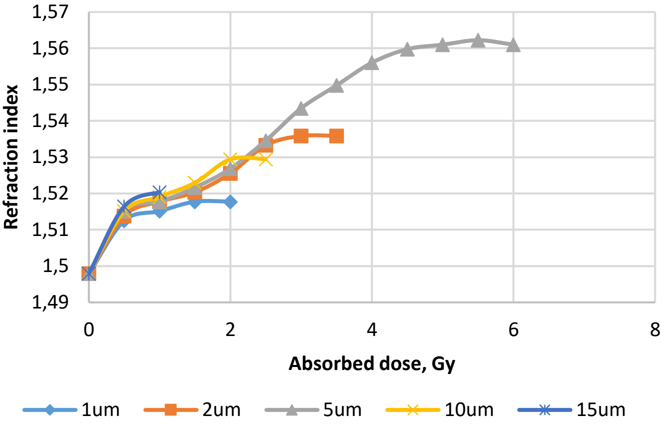


Figure 27. Dependence of refractive index on absorbed dose of VIPET gel

Obviously, due to the higher physical and optical densities, the absolute values of nPAG dosimetric gel refractive index differ from the VIPET dosimetric gel, but the dependency profiles are similar.

In this experiment, the main measured parameter was the change in sensor response (change in the SPR observation light incidence angle) that was used to calculate the irradiated dosimetric gel refractive index. Figures 26 and 27 show that dependency between gel layer thickness and maximum measurable dose is not direct, indicating that there is an optimal amount dosimetric gel for effective dose registration. With small dosimetric gel layer thicknesses, gel layer is fully polymerized even at low absorbed doses of 0.5-1 Gy. SPR signal registration remains effective for larger gel thicknesses, but after reaching a certain point (after reaching 5 <x <10µm layer thickness in these experiments), maximum recordable dose drops due to photon dispersion and absorption processes in the gel (Fig. 26 and Fig. 27).

It has been found that in these plasmonic dosimetry sensor functionalized with dosimetric gel, there is a gel thickness (5µm) at which the signal can be recorded with the highest dynamic range. Based on the obtained results (Fig. 28), highest measurable dose using plasmonic dosimetry sensor with a gel (nPAG or VIPET) layer thickness of 5µm is close to 5 Gy.

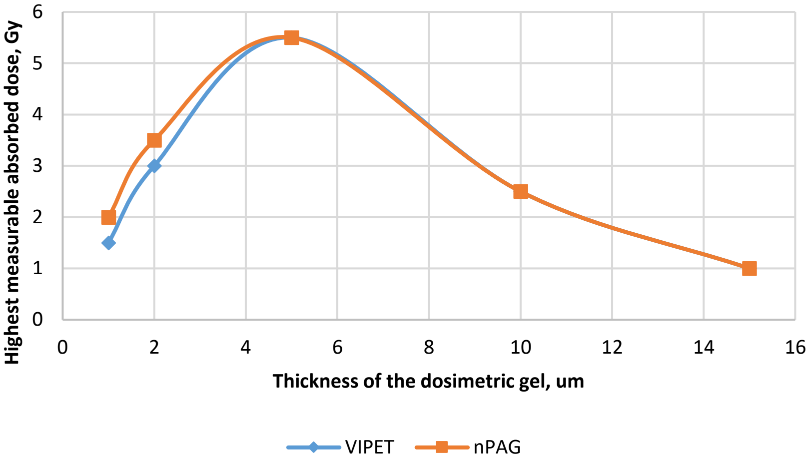


Figure 28. Dependence of highest measurable dose on the thickness of VIPET and nPAG dosimetric gel layer

3.3. Dosimetric Estimate of the Plasmonic Dosimeters

Sensitivity of this plasmonic dosimetric sensor was evaluated by analyzing dose response signal intensities' highest changes recorded at different doses in irradiated gels (Fig. 29 and Fig. 30).

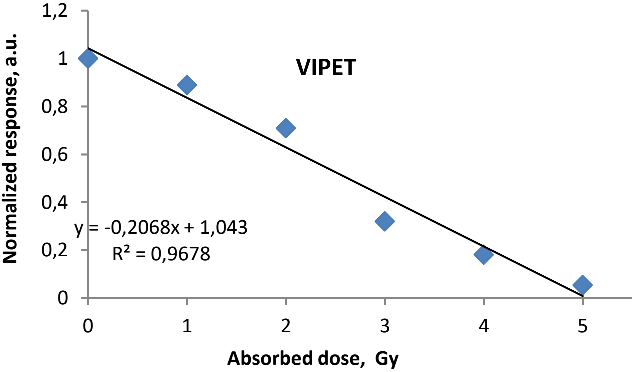


Figure 29. Dosimetric sensitivity of SPR method with VIPET gel

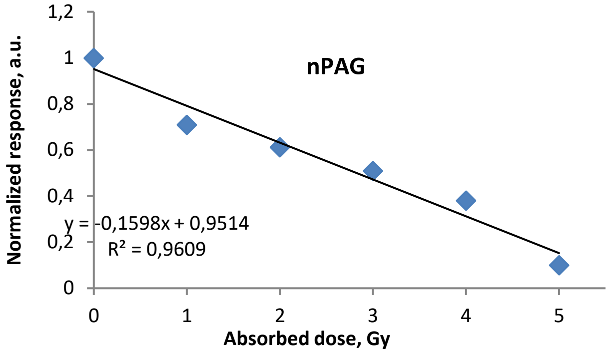


Figure 30. Dosimetric sensitivity of SPR method with nPAG gel

Plasmonic sensor with VIPET gel has been found to be more sensitive compared to plasmonic sensor with nPAG dosimetric gel. In addition, it should be emphasized that the dosimetric sensitivity of the plasmonic method with the VIPET gel (0.21 Gy^{-1}) was ~ 16% higher than the dosimetric sensitivity of the optical methods discussed in the previous section (0.18 Gy^{-1}) and literature [12, 30]. Dosimetric sensitivity of the plasmonic sensor with nPAG dosimetric gel (0.16 Gy^{-1}) was almost twice the sensitivity when compared to other methods.

CONCLUSIONS

1. The effect of exposure to low energy X-ray photons on polymerization processes in nPAG and VIPET dosimetric gels consisting of monomer and crosslinker (*N*, *N*-methylene-*bis*-acrylamide) was investigated. Change in gel optical properties and surface structures caused by these processes was analyzed. 120 keV energy was found to be sufficient to initiate the polymerization process. There are visible surface changes in the dosimetric gels with the increased dose absorbed.
2. Mathematical modeling and experimental measurements showed that the diffraction gratings of commercial optical information media (CD, DVD, HD-DVD, Blu-ray) meet the conditions required for surface plasmon resonance excitation. The concept of plasmonic dosimetric sensor has been proposed and implemented, by using discussed diffractive structures functionalized with dosimetric gels for low energy (<250 keV) absorbed dose measurements
3. A specialized open source measurement system for plasmonic dosimetric sensors' readout has been developed, which allows to record the response signal of plasmonic dosimetric sensors in angular modulation mode, both in real time and by sensor scanning after exposure. Systems' angular resolution 0.1^{o}; scanning frequency 0.25 Hz.
4. 5 μm dosimetric gel layer in the construction of the plasmonic dosimetric sensor ensures the highest dynamic measurement range with a maximum measurable dose of 5.0 Gy for both, nPAG and VIPET gels. Lowest measurable dose for nPAG dosimetric gel is 0.2 Gy, and for VIPET dosimetric gel 0.1 Gy. Resolution in both cases is 0.1 Gy.
5. Dose verification in a clinical setting showed that the-sensitivity to low-energy photons increased from 0.09 Gy^{-1} to 0.16 Gy^{-1} for nPAG dosimetric gel, and from 0,18 Gy^{-1} to 0,21 Gy^{-1} for VIPET dosimetric gel when compared to other known registration methods, in the dose range of up to 5 Gy.

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APŠVITOS DOZĖMS REGISTRUOTI SKIRTŲ PLAZMONINIŲ JUTIKLIŲ SU POLIMERINIAIS GELIAIS KŪRIMAS IR CHARAKTERIZAVIMAS

SANTRAUKA

Šiuolaikinės spindulinės medicinos esmę sudaro energijos perdavimo ir sugerties biologiniame audinyje procesai, kurie apsprendžia jonizuojančiosios spinduliuotės panaudojimą ligų diagnostikai ir onkologinių susirgimų gydymui. Atsižvelgiant į tai, kad spindulinė apšvita visada siejama su radiobiologiniais efektais, privaloma įvertinti net ir mažiausias spinduliuotės dozes. Teoriniam sąveikos procesų vertinimui bei procedūrų planavimui dažniausiai naudojamas modeliavimas, tačiau poveikio kontrolei yra būtinas eksperimentinis vertinimas, kuris atliekamas dozimetrų pagalba. Dozimetrai yra gaminami iš spinduliuotei jautrių medžiagų, kurių fizinės bei cheminės savybės kinta dėl spinduliuotės poveikio. Tarp didelės dozimetrų įvairovės (jonizacinės kameros, filmai/savaryškiai filmai, optiškai stimuliuojami (OSL), termoliuminescenciniai (TLD), cheminiai, puslaidininkiniai, puslaidininkiniai dozimetrai), dozimetriniai geliai pasižymi išskirtinėmis savybėmis, nes leidžia plačiame energijų intervale (nuo keliasdešimt keV iki keliasdešimt MeV) tiksliai ir su gera erdvine skyra registruoti trimatį dozės pasiskirstymą apšvitintame tūryje. Dozimetrinį gelį sudaro želatinos matrica bei vienas ar keli monomerai, kurių polimerizacijos laipsnis, paveikus jonizuojančiąja spinduliuote, priklauso nuo sugertosios dozės. Spinduliuotės sąlygoti struktūriniai pokyčiai dozimetriniuose geliuose siejami su gelių cheminių bei fizikinių charakteristikų pokyčiais, kurių registracijai naudojami branduolių magnetinio rezonanso (BMR) vaizdinimo, rentgeno spindulių bei optinės kompiuterinės tomografijos (OKT), optinio skenavimo, sonografijos bei kiti metodai. Nors BMR vaizdinimo metodas, vertinant dozes apšvitintuose dozimetriniuose geliuose, laikomas „aukso standartu“ didelės energijos (MeV) spindulinėje terapijoje, jis yra sudėtingas, brangus ir dėl didelių pacientų srautų prieinamas ne kiekvienoje ligoninėje.

Apkreiptinas dėmesys, jog paviršinių navikų/odos vėžio atveju naudojami mažesnių energijų (iki kelių šimtų keV) dalelių ar fotonų srautai, todėl ypač svarbu registruoti lokalias paviršiaus dozes. Paprastai šiam tikslui naudojami „taškiniai“ TLD arba OSL dozimetrai ir dozių skaitytuvai, tačiau jie yra sąlyginai brangūs, yra specifinių apribojimų registruojant > 1,0 Gy dozes. Be to, uždėjus dozimetrą ant paciento odos, dozimetras registruoja ir nuo paciento išsklaidytas daleles. Minėtus dozimetrus sėkmingai galėtų pakeisti paviršiaus plazmonų rezonanso (PPR) reiškinio pagrindu veikiantys jutikliai, suformuoti iš spinduliuotei jautrių polimerinių gelių ant difrakcinės gardelės paviršiaus.

Plazmoninių jutiklių veikimo principas yra paremtas stiprių laisvųjų elektronų osciliacijų sužadinimu ribiniame sluoksnyje tarp metalo ir dielektriko, naudojant poliarizuotą šviesą. Esant pasirinktam charakteringam spindulio kritimo kampui

ar bangos ilgiui, nuo metalo-dielektriko ribos atspindėtos šviesos intensyvumo pokyčiai leidžia charakterizuoti dielektrinio sluoksnio chemines bei fizikines savybes. plazmoniniai jutikliai dažniausiai taikomi biotechnologijose, kur vertinamos įvairių organinių junginių koncentracijos ir reakcijų greitis [6-8]. PPR reiškinio pritaikymo pavyzdžių spinduliuotės dozėms registruoti rasti literatūroje nepavyko.

Įvertinus žinomų plazmoninių jutiklių charakteristikas, dozimetrinių gelių lygiavertiškumą biologiniam audiniui ir perspektyvas kurti specifinės paskirties didelio jautrumo jonizuojančiosios spinduliuotės poveikiui gelius ir galimybę realizuoti tiesioginį paviršinės dozės registravimą (be atgalinės sklaidos komponentės) šiame darbe buvo pasiūlyta ir realizuota gelinio PPR dozimetro koncepcija.

Darbe pristatomas mažų energijų (<250 keV) rentgeno fotonų spinduliuotei jautrių dozimetrinių gelių iš akrilamido (AAm) ir N,N-metilen-bis-akrilamido (BIS) bei vinilpirolidono ir BIS formavimas ir jų fizinių bei cheminių savybių tyrimas bei šių gelių panaudojimas, konstruojant plazmoninius dozimetrus, skirtus mažų dozių (<10 Gy) registravimui. PPR reiškinio sužadinimui pasirinktas difrakcinės gardelės metodas, o dozių vertinimui, naudojant plazmoninį dozimetrą, sukurta originali dozimetrijos sistema.

Disertacijos tikslas ir uždaviniai

Disertacijos tikslas – sukurti ir charakterizuoti plazmoninius jutiklius su polimeriniais geliais ir juos pritaikyti mažų, jonizuojančios spinduliuotės, dozių registravimui

Šiam tikslui įgyvendinti iškelti šie uždaviniai:

- Ištirti nedidelių energijų (<250 keV) fotonų inicijuotų polimerizacijos procesų geliuose su tinklikliu sąlygotus optinių savybių pokyčius.
- Įvertinti paviršiaus plazmonų rezonanso sužadinimo galimybes optinių informacijos laikmenų difrakcinėse struktūrose ir jas funkcionalizuoti tirtais dozimetriniais geliais.
- Sukurti plazmoninių jutiklių nuskaitymo sistemos modelį ir jį pritaikyti
- medicininių apšvitos dozių vertinimui.
- Verifikuoti dozių registravimą, naudojant plazmoninius jutiklius.

IŠVADOS

1. Ištirtas apšvitos mažų energijų rentgeno fotonais poveikis polimerizacijos procesams nPAG ir VIPET dozimetriniuose geliuose, sudarytuose iš monomero ir tinkliklio (*N*,*N*-metilen-*bis*-akrilamido), bei išanalizuota šių procesų sąlygota gelių optinių bei paviršiaus struktūrų kaita. Nustatyta, kad 120 keV energijos pakanka polimerizacijos procesui inicijuoti. Matomi paviršiniai dozimetrinių gelių pokyčiai didėjant sugertajai dozei.
2. Matematinis modeliavimas ir eksperimentiniai matavimai, parodė, kad komercinių optinių informacijos laikmenų (CD, DVD, HD-DVD, Blu-ray) difrakcinių gardelių periodai ir gyliai tenkina paviršiaus plazmonų rezonanso sužadinimo sąlygas. Pasiūlyta ir įgyvendinta plazmoninio dozimetrinio jutiklio, koncepcija, įveiklinant dozimetriniais geliais funkcionalizuotas aptartas difrakcines struktūras mažos energijos (<250 keV) jonizuojančios spinduliuotės dozėms registruoti.
3. Sukurta specializuota atviro kodo principu veikianti dozimetrinių plazmoninių jutiklių nuskaitymo sistema, leidžianti registruoti plazmoninių dozimetrinių jutiklių atsako signalą kampinės moduliacijos režime, tiek realiu laiku, tiek atliekant jutiklio nuskaitymą po apšvitos. Sistemos kampinė skyra 0,1°; nuskaitymo dažnis - 0,25 Hz.
4. 5 µm storio gelio sluoksnis plazmoninio dozimetrinio jutiklio konstrukcijoje užtikrina plačiausią darbinių dozių ruožą su maksimalia išmatuojama 5,0 Gy doze nPAG ir VIPET geliams. Mažiausios išmatuojamos dozės vertė nPAG geliui - 0,2 Gy, o VIPET geliui - 0,1 Gy, esant dozės skyrai 0,1 Gy.
5. Dozių registravimo verifikacija klinikinėje aplinkoje parodė, kad lyginant PPR metodą su kitais žinomais registravimo metodais, dozės registravimo jautris mažų energijų fotonais apšvitintam nPAG geliui padidėjo nuo 0,09 Gy^{-1} iki 0,16 Gy^{-1}, o VIPET geliui - nuo 0,18 Gy^{-1} iki 0,21 Gy^{-1}, vykdant dozių registraciją iki 5 Gy.

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