RESULTS OF STUDY THE INFLUENCE ELECTRON IRRADIATION ON THE HIGH-FREQUENCY DIELECTRIC PROPERTIES OF ZIRCONIA NANOCERAMICS

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The use of dielectric materials in accelerator technology makes it necessary to research the change of dielectric constant and dielectric loss under relativistic electrons irradiation. The experiments were performed on electron linac LUE-40 with electron energy up to 100 MeV. Special attention was attends to RF measurements of small permittivity changes. It was found that the permittivity of zirconia nanoceramics (ZrO_2 *– 4 wt% MgO*) was decreased on average $(0.23 \pm 0.02)\%$ as a result of irradiation by electron beam with energy 41 MeV and the fluence of $\approx 10^{18}$ cm^{-2}.

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INTRODUCTION

The dielectric materials are widely used in accelerating technique. For the past time the new dielectric based accelerating structures were designed and experimentally researched [1 - 4]. The zirconia nanoceramics which developed and manufactured at the DonPhTI [5, 6] can be used in RF-resonators of the multi-bunch accelerator [4]. It necessitate detailed study the dielectric properties of this material in the S-band of electromagnetic waves. First of all it concerns to the real part of complex permittivity ε and loss tangent tgδ.

To the moment of our research beginning the unambiguous information about dielectric properties of zirconia ceramics was absent in literature. The measurement results for the same materials substantially differ from each other [7]. Undoubtedly, it is first of all connected with the distinction of technology of making the ceramics, its composition and crystalline structure.

The permittivity change of dielectric accelerating structure during exploitation can lead to the undesirable change of beam parameters. The basic possible source of dielectric properties change during the electron, neutron and gamma irradiation is radiation-induced defects of crystalline structure [8]. It is known that appearance of the radiation-induced defects lead to the increase of conductivity and, as a result, to the increase of dielectric losses. As concerns permittivity behavior, there are a few physical mechanisms that can result both in an increase and to reduction of permittivity. Under irradiation of dielectrics by neutrons or high energy electrons as a result of nuclear reactions new elements appear in material. The atom concentration of these elements even at the irradiation with large fluence is small. But it is unknown how they effect on the dielectric properties. The change of dielectric properties is also possible in case the crystalline structure change under the irradiation action. This question requires the detailed study as well.

The dielectric properties of ceramic material depend on its composition, density, technology of making. Therefore, appropriate studies should be carried out in each case. In our case the research of permittivity change after an irradiation has some peculiarities. Firstly, after irradiation the samples have a high level of induced radiation that does not allow to conduct RF measuring in ordinary laboratory conditions during a long time (to 1 year to and more). In this connection the mass of a sample must be small. It is known, also, that the permittivity of zirconia ceramics is high and lies within the limits of 17…27. These features demanded the careful analysis and selection the methods for permittivity and dielectric losses measuring.

1. METHODS OF PERMITTIVITY AND LOSS TANGENT MEASURING

Presently there are many methods of complex permittivity determination, based on different principles. The resonant methods are the most widespread in RF range. The basic idea of these methods consists in the measurement of the eigenfrequency shift and quality factor shift of a resonator with and without a dielectric sample. Then, on the base of experimental data, analytically or with numerical method we can to find the permittivity. We will consider some realizations of resonant method bellow.

The use of methods based on small perturbation approach allows to measure the complex permittivity of small size samples. The approach of small perturbations supposes that the dielectric sample placed in the resonator disturb slightly the electromagnetic field. The error of measuring is grows with increasing the geometrical sizes and permittivity. Therefore applicability of this method in the S band is practically limited to the value of permittivity ε=7. These methods can be used only for simplest resonators that have the exact analytical solution of eigenfrequency problem. So, in our case this method is inapplicable.

With the computing engineering progress and appearance of the computer codes that determine eigenfrequencies and quality factor of free-form resonators, the new approach to complex permittivity measurement has been developed, see for example [9]. It gives an opportunity to investigate the dielectric samples of free-form and sizes, to take into account influence of auxiliary elements of resonator (for example, holders of sample and RF signals connectors). We have chosen this method with the partial filling of resonator with dielectric for the preliminary measuring of zirconia ceramics permittivity and its change due to the electron irradiation. The error in measurement permittivity is determined by the following basic factors:

- error in measuring resonant frequency – mainly caused by inaccuracy at assembling - disassembling of resonator and sample setting;
- numeral model adequacy to the experiment conditions (sizes of resonator and sample, sizes and properties of holder);
- errors of simulation.

If a dielectric sample has small RF losses it is possible to use the method of the complete filling of the cavity. Thus it is possible to attain a maximum sensitivity for measurement a change of permittivity. Really, for a cylindrical resonator an E_{010} eigenfrequency and a quality factor are expressed as follows:

$$f_0 = \frac{c \cdot p_{01}}{2\pi R \cdot \sqrt{\varepsilon}}, \quad (1)$$

$$Q_0 = \frac{1}{\frac{1}{Q_m} + tg\delta}. \quad (2)$$

Here R is the radius of resonator, p_{01} is the first root of Bessel function of a zero order, ε – relative permittivity of dielectric that fills a resonator fully, Q_m is quality factor of resonator without dielectric. From (1) it follows, that, for example, in an S band, the change of 1% in ε causes the change of 10^7 Hz in eigenfrequency.

1.1. RESONATORS FOR PERMITTIVITY MEASURING

For the preliminary measuring the cylindrical resonators have been designed and fabricated in DonPhTI and in NSC KIPT. The investigated zirconia samples of *ZrO_2 – 4 wt% MgO* ceramics are disks with a diameter about 10 mm and a high about 2.5 mm.

The resonator fabricated in NSC KIPT is shown in Fig. 1. The ceramics sample has been set on the teflon holder. Frequency of E_{010} mode and quality factor without a sample are 2700.9 MHz and $(11.7\pm0.3)\cdot10^3$. The setting of sample changes the resonant frequency on 5.5...7.0 MHz. The relationship between this frequency change and sample permittivity have been definite by a numeral design with code SUPERFISH [10]. From the calculations it follows, that for this sample (if ε≈20) $\Delta f_0/\Delta\varepsilon \approx 7$ kHz/%. We used the special device to stable of RF contacts and statistical treatment of measuring results. It decreased the error in measuring of resonant frequency, which is caused by resonator and sample mounting to ±60 kHz. The measurements conducted both in NSC KIPT and DonPhTI shows that the permittivity of zirconia samples before irradiation is 23±2, and tgδ does not exceed $3\cdot10^{-3}$.

The ceramics samples for permittivity measurement must to have a regular form and must be processed with high precision. The thickness of a disk sample must be uniform with accuracy of a few microns. The same requirements are imposed on the disk diameter. Unfortunately, our samples of zirconia ceramics are not satisfied such requirements. The inaccuracy of processing both disk thickness in different points and disk diameter was 10…150 microns. Therefore it is impossible to exactly reproduce the complex shape of samples for simulation. It limited the accuracy of permittivity measuring with the value of 10%. Thus, evidently, that for measuring of small changes of permittivity (less than 1%) it is necessary to use another method.

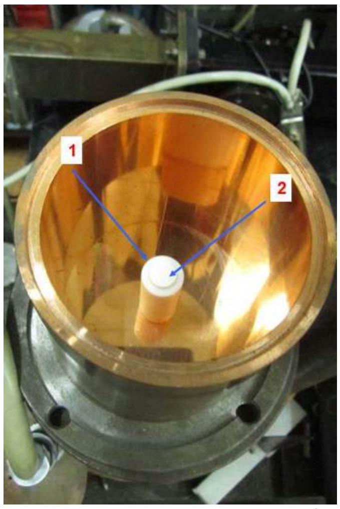

Fig. 1. Sample (2) in the cavity on the teflon holder (1)

The resonance method with the complete filling of the cavity with dielectric can be an alternative approach. In this case permittivity and loss tangent can be found from exact analytical expressions. Obviously, this method is applicable in the case of small dielectric losses. If to suppose that $tg\delta=5\cdot10^{-3}$, the quality factor of such resonator will be about 200. It is enough for measuring of all resonator characteristics. To eliminate the errors related to the gaps between dielectric and metal, it is necessary to coat metal layer on the surface of the investigated dielectric sample. Thickness of the coating must be substantially more than a skin depth. The skin depth for copper and silver in a 10-cm range approximately equals 1.2 μm.

For realization of this method new samples have been fabricated. The basic dielectric disks sizes were processed with accuracy of 10 microns. If the diameter of resonator is 20 mm, height is 3 mm and permittivity is 21, the E_{010} mode frequency is about 2500 MHz. The change of ceramics permittivity from 21 to 20 (5%) changes the E_{010} eigenfrequency at 40 MHz.

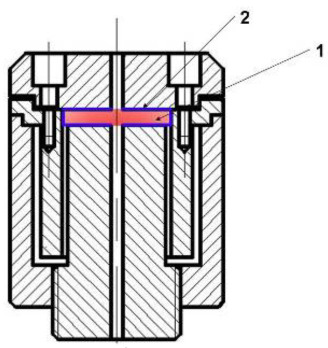

Fig. 2. The device for measuring microwave characteristics of ceramic samples. 1 – sample (R = 10 mm; h = 3 mm); 2 – metal coating

For permittivity measuring the special device has been designed and created (Fig. 2). In case of necessity it allows to conduct the measuring of samples not coated by metal. RF measuring of resonant frequency and quality factor has been conducted with schema presented in Fig. 3.

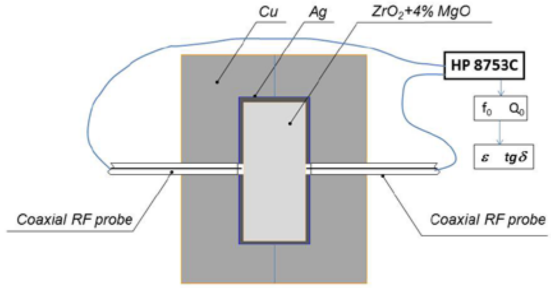

Fig. 3. Measurement scheme

We tested different methods for coating metal on zirconia ceramic dicks: magnetron spraying and thermal spraying of copper, deposition of silver by brazing method. The best results on durability of coating have been obtained with silver (Fig. 4). The thickness of the coating was 8 µm, which is much more than the skin depth.

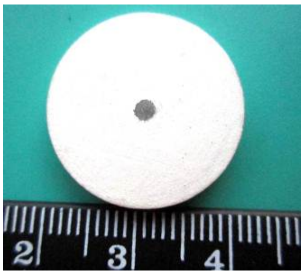

Fig. 4. RF resonator – sample coated with silver. The central hole in the center is designed for RF excitation the resonator

The calculation of cavity frequency and its quality factor vs. the value ε and tgδ has been conducted by SUPERFISH. These dependences for samples #1 are presented in Figs. 5, 6. It can be seen that 1% change in permittivity (ε≅25) results to frequency change of 11.82 MHz. The dependencies of E_{010} eigenfrequency vs. the radius and the height of resonator have been obtained – they are 230 and 4 MHz/mm accordingly. The accuracy of radius measuring is ±10 µm, therefore the error of determination the resonant frequency is ±2.3 MHz and the error of permittivity determination is ±0.2%. The tgδ for the fully filled resonator is determined from the expression (2). The quality factor Q_m in our case depends on the coating quality and roughness of the sample surface. We shall notice that the relative change of tgδ and ε for the same sample before and after the irradiation is only determined by the change of dielectric properties.

Quality factor measuring on five samples with different silver coverage has shown that Q_0 value lays within the limits of 280…335, i.e. tgδ = (2.4…2.9)·10^{-3}. These results are consistent with measuring, that has been conducted in the resonator (see Fig. 2) with the dielectric dicks not coated by silver – tgδ ≅ (2.1…2.5)·10^{-3}. For electron beam irradiation two samples with the best quality of coating have been chosen.

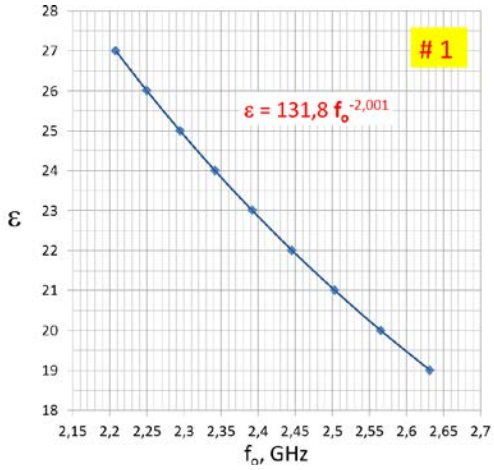

Fig. 5.

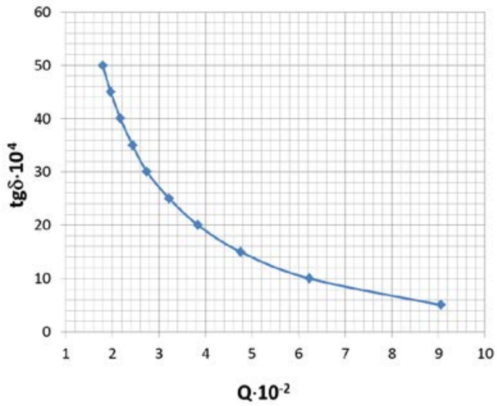

Fig. 6.

The E_{010} eigenfrequency and quality factor of the samples were measured immediately before irradiation. Measurements were carried out at a temperature of (18±0.5)°C. Each parameter was measured at least ten times with assembling – disassembling procedure the measuring device. The results were subjected to statistical processing. The measurement results are summarized as follows:

sample #1 f_0 = 2281.79±0.12 MHz, Q_0=335±10;

sample #2 f_0 = 2275.79±0.12 MHz, Q_0=303±10.

Based on the example data one can determine the dielectric constant and loss tangent of each sample. For sample # 1 ε= 25.29 ±0.05 and tgδ= 2.4·10^{-3}. For sample #2 ε = 25.36±0.05 and tgδ= 2.8·10^{-3}.

2. ELECTRON IRRADIATION AND THE DIELECTRIC PROPERTIES OF ZIRCONIA NANOCERAMICS

Experiments with electron irradiation were conducted on the linac LUE-40 [11]. At the first phase irradiation of ceramic discs with a diameter of 10 mm and a thickness of 2.5 mm was conducted. There were about 20 samples irradiated with different electron beam fluences (from 2·10^{16} to 2·10^{18}cm^{-2}) and the electrons energy to 90 MeV. The change in temperature of samples during irradiation did not exceed 100°C. Research of irradiated samples allowed getting the information about formation of radioactive isotopes, time dependence induced radioactivity and formation of radiation defects [12]. It is revealed also, that krypton atoms are centers of segregation of point defects.

The ceramic samples after irradiation were dark gray color (Fig. 7), which is likely due to the formation of oxygen vacancies excess in zirconia.

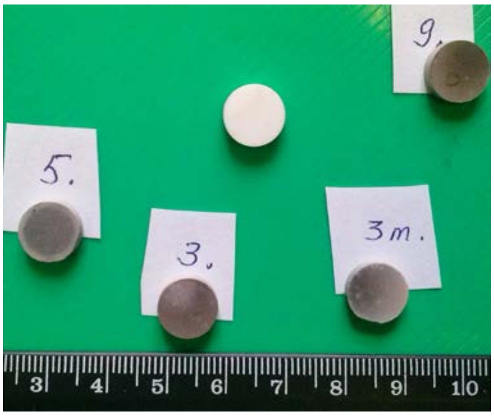

Fig. 7. Samples ZrO_2 – 4 wt% MgO (№№ 5, 3, 3m, 9) after electron irradiation. In the center of the picture – not irradiated sample

Change the color of zirconia samples under the influence of temperature and vacuum or neutral atmosphere is known fact. It is based on breach of stoichiometry (loss of oxygen). The reduction of oxygen in the zirconia formula from 2 to 1.96 is sufficient for such change in color. After irradiation ceramic samples were annealed at 500°C. The samples color returned to the original state, i.e. the excess oxygen vacancies were annealed and stoichiometric composition was restored. This confirms the assumption that under electron irradiation in zirconia lattice oxygen vacancies are formed.

Research of the dielectric properties change of ceramic samples with a diameter of 10 mm was carried out by using the cavity that depicted in Fig. 1. The experimental data do not allow to draw a definite conclusion about the influence of irradiation on the permittivity because of the low accuracy of measurements (error to 10%). Therefore, further detailed study was conducted with samples of 20 mm, using the method of complete filling resonator.

The irradiation of 20 mm samples has been conducted on the linac exit with electrons energy of 41 MeV and average current of 4.54 μA. The irradiation time for each of two samples was 16.5 hour. Thus, a common amount of particles was $1.68\cdot10^{18}$. Samples have been placed in the special radiator for cooling. The temperature was measured with thermocouple. As the thermocouple must not be exposed by beam, it has been set in the distance of 3 mm from the samples border. The temperature in the sample center has been determined by simulation. According to the results of simulation, the difference between the thermocouple indication and temperature in the sample center does not exceed 100°C. During all irradiation time the temperature measured by a thermocouple was less than ≈70°C (i.e. less than 170°C in the sample center). To obtain the necessary electrons density distribution on a sample, a diffuser (an aluminum plate 1.5 mm thick) has been set after the linac exit.

After the irradiation the samples have had a high level of activity. After «cooling» during a month the repeated high-frequency measuring have been conducted. The measurement conditions were identical with those measurements before irradiation. During the measuring the following results have been obtained: sample #1 f_0 =2284.52±0.12 MHz, Q_0=337±10; sample #2: f_0 =2278.42±0.2 MHz, Q_0=305±10.

Thus, comparing the results of measuring of eigenfrequencies and quality factor of resonators before and after irradiation, it is possible to draw the following conclusions.

As a result of influence of electron beam irradiation with the fluence of $\approx10^{18}$cm^{-2} the frequency of E_{010} mode increased for a sample #1 and sample #2 by 2.73±0.12 and 2.76±0.2 MHz accordingly. This frequency change can be explained by a decrease in the dielectric constant of zirconia nanoceramics (*ZrO_2 – 4 wt% MgO*) on average (0.23±0.02)%. Electron beam irradiation did not lead to a change in the dielectric loss tangent (within range of ±3%). Causes of changes in permittivity of zirconia nanoceramics due to high energy electron irradiation will be the subject of our future work.

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РЕЗУЛЬТАТЫ ИССЛЕДОВАНИЯ ВЛИЯНИЯ ЭЛЕКТРОННОГО ОБЛУЧЕНИЯ НА ВЫСОКОЧАСТОТНЫЕ ДИЭЛЕКТРИЧЕСКИЕ СВОЙСТВА ЦИРКОНИЕВОЙ НАНОКЕРАМИКИ

Н.И. Айзацкий, В.А. Бочаров, Н.П. Дикий, А.Н. Довбня, В.А. Кушнир, В.В. Митроченко, О.Д. Никитина, И.Н. Онищенко, Л.И. Селиванов, Д.Л. Степин, С.А. Пережогин, И.А. Даниленко, Т.Е. Константинова, В.Ф. Жигло

Использование диэлектрических материалов в ускорительной технике делает необходимым проведение исследований процессов изменения диэлектрической проницаемости и диэлектрических потерь при облучении релятивистскими электронами. Приводятся результаты экспериментального изучения диэлектрических свойств циркониевой нанокерамики после облучения высокоэнергетическими электронами. Эксперименты проводились на ускорителе электронов ЛУЭ-40 с энергией частиц до 100 МэВ. Особое внимание уделяется методам измерения на СВЧ малых изменений диэлектрической проницаемости. Установлено, что в результате воздействия электронного пучка с энергией 41 МэВ и флюенсом $\approx 10^{18}$см$^{-2}$ диэлектрическая проницаемость циркониевой нанокерамики (ZrO_2 – *4 об.%* MgO) уменьшается в среднем на (0,23 ± 0,02)%.

РЕЗУЛЬТАТИ ДОСЛІДЖЕННЯ ВПЛИВУ ЕЛЕКТРОННОГО ОПРОМІНЕННЯ НА ВИСОКОЧАСТОТНІ ДІЕЛЕКТРИЧНІ ВЛАСТИВОСТІ ЦИРКОНІЄВОЇ НАНОКЕРАМІКИ

М.І. Айзацький, В.А. Бочаров, М.П. Дикий, А.М. Довбня, В.А. Кушнір, В.В. Митроченко, О.Д. Нікітіна, І.М. Оніщенко, Л.І. Селіванов, Д.Л. Стьопін, С.А. Пережогін, І.А. Даниленко, Т.Є. Константинова, В.Ф. Жигло

Використання діелектричних матеріалів у прискорювальній техніці робить необхідним проведення досліджень процесів зміни діелектричної проникності та діелектричних втрат при опроміненні релятивістськими електронами. Наведено результати експериментального вивчення діелектричних властивостей цирконієвої нанокераміки після опромінення високоенергетичними електронами. Експерименти проводилися на прискорювачі електронів ЛУЕ-40 з енергією частинок до 100 МеВ. Особлива увага приділяється методам вимірювання на НВЧ малих змін діелектричної проникності. Встановлено, що в результаті впливу електронного пучка з енергією 41 МеВ і флюенсом $\approx 10^{18}$см$^{-2}$ діелектрична проникність цирконієвої нанокераміки (ZrO_2 – *4 об.%* MgO), зменшується в середньому на (0,23 ± 0,02)%.