

## SCIENTIFIC CONTRIBUTIONS

in the presented by Chief Assistant Professor Dr. Stanislav Slavov  
publications, patents and patent applications

for participation in the competition for an associate professor in Mathematical Modeling and  
Application of Mathematics (mathematical analysis of the structure of condensed matter in French)  
(SG, issue 67 of 13.08.2021)

### I. Major scientific contributions.

#### I.1 Synthesis, structure and microstructure of new materials.

1. Synthesis, structure and microstructure of bismuth-titanate ceramics, glass-ceramics and glasses from the system  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2\text{-Nd}_2\text{O}_3$ .
2. Synthesis and microstructure of compositions in the system  $\text{La}_2\text{O}_3\text{-Gd}_2\text{O}_3\text{-PbO-MnO-B}_2\text{O}_3$ .
3. Synthesis and microstructure of compositions in the system  $\text{TeO}_2\text{-Bi}_2\text{O}_3\text{-Nb}_2\text{O}_5\text{-ZnO}$ .
4. Synthesis and microstructure of new graphene oxide composites, in combination with nanoscale ZnO.

#### I.2. Dielectric characteristics of ceramics, glass-ceramics and glasses.

1. Dielectric characteristics of new materials in the system  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2\text{-Nd}_2\text{O}_3$ .
2. Dielectric characteristics of new materials in the system  $\text{La}_2\text{O}_3\text{-Gd}_2\text{O}_3\text{-PbO-Mn}_2\text{O}_3\text{-B}_2\text{O}_3$ .
3. Dielectric characteristics of new graphene oxide composites, in combination with nanoscale ZnO.

#### I.3. Mathematical modeling and algorithms for parametric estimation.

1. Methods for estimating the parameters of differential equations.
2. Models created on the basis of graph theory for calculating the fractal dimension of 3D objects for the materials science.

### II. Scientific and applied contributions

1. New constructions and materials for ceramic capacitor batteries.
2. New composite materials from natural raw materials and method for their industrial production.

## **I. Major scientific contributions.**

### **I.1. Synthesis, structure and microstructure of new materials.**

#### **1. Synthesis, structure and microstructure of bismuth-titanate ceramics, glass-ceramics and glasses from the system $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2\text{-Nd}_2\text{O}_3$ .**

- (1.1.) Polyphase samples were synthesized in the system  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-Nd}_2\text{O}_3$  using the method of melt quenching, and developed a methodology for the control of the constituent material Aurivillius and pyrochlore crystalline phases. The method is used to control the high-frequency dielectric characteristics [II.1.].
- (1.2.) Glass-crystal materials for sensors in the system  $\text{SiO}_2\text{-Bi}_2\text{O}_3\text{-TiO}_2$  have been synthesized by a method of melt quenching and controlled crystallization of the glass and subsequent synthesis of thin layers by ink-screen-printing technique [I.2.].
- (1.3.) The synthesis of selected samples from the system  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2\text{-Nd}_2\text{O}_3$  is made on the basis of freely cooled melt (from 1450 °C and 1100 °C to room temperature). The method of the initial control of the amount of starting compositions ensures the formation of polyphase glass-ceramics with content-controlled crystalline phases  $\text{Bi}_{12}\text{TiO}_{20}$  и  $\text{Bi}_4\text{Ti}_3\text{O}_{12}$  [I.3.] and single-phase polycrystalline glass-ceramic material contains  $\text{Bi}_{12}\text{TiO}_{20}$  [I.4.].
- (1.4.) The influence of  $\text{SiO}_2$  и  $\text{Nd}_2\text{O}_3$  on glass formation in bismuth titanates has been studied [I.7.]. The replacement of  $\text{Bi}_2\text{O}_3$  with  $\text{Nd}_2\text{O}_3$  to 10 mol% leads to obtaining a complex of multicomponent glasses and glass-crystal materials with high thermal stability. When adding  $\text{SiO}_2$  and  $\text{Nd}_2\text{O}_3$  have also been shown to help control crystallization and synthesis temperature over a wide concentration range [II.3.], [I.10.].
- (1.5.) A new approach for the synthesis of ferroelectric ceramic materials from the system has been implemented  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2\text{-Nd}_2\text{O}$  based on mixing the precursors in a powerful ultrasonic field generated by a specially designed device [I.9.]. In this approach, sonosynthesis stimulates the priority formation of crystal phases, which are characterized by lower synthesis temperatures. In the glasses obtained after subsequent thermal synthesis, a high content of  $\text{TiO}_2$ .
- (1.6.) The area of glass formation in a system  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$  is defined at cooling rate  $10^2$  K/s [I.5., II.6.]. Thermostable glasses are available in the range 10-50 mol %  $\text{SiO}_2$ , from 10 to 50 mol %  $\text{Bi}_2\text{O}_3$  and from 10 to 50 mol %  $\text{TiO}_2$ . A hypothesis for the formation of an amorphous network with a non-traditional glass-forming network with  $\text{Bi}_2\text{O}_3$ ,  $\text{TiO}_2$  and a classic glass former  $\text{SiO}_2$  is proposed.

#### **2. Synthesis and microstructure of compositions in the system $\text{La}_2\text{O}_3\text{-Gd}_2\text{O}_3\text{-PbO-MnO-B}_2\text{O}_3$ .**

- (2.1.) They are synthesized using the method of melting and quenching, materials in the system  $\text{La}_2\text{O}_3\text{-Gd}_2\text{O}_3\text{-PbO-MnO-B}_2\text{O}_3$ , in which the presence of only one perovskite crystalline phase has been established  $(\text{La}_{1-x}\text{Gd}_x)_{0.6}\text{Pb}_{0.4}\text{MnO}_3$  [I.1.]. In the system, outside the area of glass formation, glass-ceramic material is obtained directly from the supercooled melt [I.6.].

#### **3. Synthesis and microstructure of compositions in the system $\text{TeO}_2\text{-Bi}_2\text{O}_3\text{-Nb}_2\text{O}_5\text{-ZnO}$ .**

- (3.1.) Tellurium-bismuth glasses were synthesized in the system  $\text{TeO}_2\text{-Bi}_2\text{O}_3\text{-Nb}_2\text{O}_5\text{-ZnO}$ , in which the identified phases are  $\text{ZnTeO}_3$  and  $\text{TiTe}_3\text{O}_8$  and an amorphous network composed mainly of  $\text{TeO}_4$  structural groups [I.5.].

#### **4. Synthesis and microstructure of new graphene oxide composites, in combination with nanoscale ZnO.**

- (4.1.) A complete series of graphene oxide was synthesized in combination with nanosized zinc oxide, and the structural and phase characteristics of the obtained composites were made [I.8.].

## **I.2. Dielectric characteristics of ceramics, glass-ceramics and glasses.**

### **1. Dielectric characteristics of new materials in the system $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2\text{-Nd}_2\text{O}_3$ .**

- (1.1.) The dielectric properties of the samples in the system  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-Nd}_2\text{O}_3$  have been studied in frequency 2.7 GHz show dielectric constant and dielectric losses with approximately equal parameters to those of lead-containing commercial samples. The control of dielectric parameters is realized by precise control of the percentage of initial oxides and synthesis temperatures [II.1.].
- (1.2.) The dielectric characteristics of sensors based on glass crystalline materials in the system  $\text{SiO}_2\text{-Bi}_2\text{O}_3\text{-TiO}_2$  have been studied [I.2.]. The samples have very low values of electrical conductivity ( $10^{-6}\text{-}10^{-9} (\Omega\cdot\text{cm})^{-1}$ ), dielectric constant between 1000 and 3000 at room temperature. At the Curie temperature (830 °C) the dielectric constant is 180000 and 7000.
- (1.3.) Depending on the controlled melting conditions and additional heat treatment of the supercooled compositions, different polyphase glass-ceramic materials with different microstructures in the systems  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$  and  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-Nd}_2\text{O}_3$ , as well as the presence of several crystallographic phases, the conductivity is in the range  $10^{-6}\text{-}10^{-9} (\Omega\cdot\text{cm})^{-1}$  [I.10.].

### **2. Dielectric characteristics of new materials in the system $\text{La}_2\text{O}_3\text{-Gd}_2\text{O}_3\text{-PbO-Mn}_2\text{O}_3\text{-B}_2\text{O}_3$ .**

- (2.1.) The dependence of the dielectric relaxation processes for samples in the system  $\text{La}_2\text{O}_3\text{-Gd}_2\text{O}_3\text{-PbO-Mn}_2\text{O}_3\text{-B}_2\text{O}_3$  of the perovskite phases of the type  $(\text{La}_{1-x}\text{Ga}_x)_{0.6}\text{Pb}_{0.4}\text{MnO}_3$  is studied. Polyphase glasses demonstrate higher activation energy even at low relaxation times. In more complex structures, these processes are difficult due to their limited mobility [I.1.].

### **3. Dielectric characteristics of new graphene oxide composites, in combination with nanoscale ZnO.**

- (3.1.) In the studied temperature range of about 150 K to 400 K and a frequency range of  $10^2$  Hz to  $10^6$  Hz no relaxation processes were observed in the studied composite materials with the participation of graphene oxide [I.8.]. The studied composites have a behavior similar to that of dielectrics with ionic relaxation polarization, which causes increased dielectric losses, which increases with increasing temperature, ie the characteristic maximum for polar dielectrics is missing in the dependence  $\text{tg}\delta$  (T).

## **I.3. Mathematical modeling and algorithms for parametric estimation.**

### **1. Methods for estimating the parameters of differential equations.**

- (1.1.) The iteration scheme Picard-Lindelöf has been modified to show an iterative algorithm for estimating the parameters of ordinary differential equations. The algorithm, in addition to inheriting the advantages shown in the classical algorithms, the parameters can be transformed into a form more convenient for calculation.

### **2. Models created on the basis of graph theory for calculating the fractal dimension of 3D objects for the materials science.**

- (2.1.) A new approach to model recognition (crystallographic structures, grain size) using graph theory and its application in mechanical engineering has been developed [II.5.]. The estimation method is based on calculating the fractal dimension of 3D objects and calculating the density of 3D visibility graphs at a given SEM image.

## **II. Scientific and applied contributions**

### **1. New constructions and materials for ceramic capacitor batteries.**

- (1.1.) A combination of ceramic and glass-ceramic materials has been created, which in one with a specific construction and a combination of conventional and new physical principles,

which are the basis of the created hybrid capacitor bank of a new type [Patent № BG67056 B1/01.06.2020].

## 2. **New composite materials from natural raw materials and method for their industrial production.**

(2.1.) A new ceramic material has been created, as well as a new technological sequence of procedures for industrial production of conductive composite ceramic materials with application for membranes for hydrogen generators. [Applied patent: RO1358 /25.05.2020 ; U/00020/26.05/2020]

### **Summaries of articles**

**[I.1.]** R Raykov, A Staneva, Y Dimitriev, S Slavov, S Soreto Teixeira, PR Prezas, L. Costa, Dielectric relaxation in glass and glass-ceramic materials of the system  $\text{La}_2\text{O}_3\text{-Gd}_2\text{O}_3\text{-PbO-MnO-B}_2\text{O}_3$ , *International Journal of Applied Glass Science* 10 (1), 75-82, 2019

- Borate-based glasses in the system  $\text{La}_2\text{O}_3\text{-Gd}_2\text{O}_3\text{-PbO-MnO}_2\text{-B}_2\text{O}_3$ , synthesized by the method of melting and quenching are synthesized with a fixed content of  $\text{B}_2\text{O}_3$  (% mol = 30). Dielectric measurements are made in the frequency range of 100 Hz to 1 MHz and temperatures from 100 °C to 400 °C. A multiphase product was obtained with  $(\text{La}_{1-x}\text{Ga}_x)_{0.6}\text{Pb}_{0.4}\text{MnO}_3$  perovskite type crystalline phase, in combination with the amorphous phase. Dielectric relaxation properties are highly phase dependent, especially when amorphous phases with high activation energy and low relaxation frequency due to dipole mobility limitations.

**[I.2.]** AS Afify, SS Slavov, AER Mahmoud, M Hassan, M Ataalla, A Staneva, Amr Mohamed, Determination of the Sensing Characteristics of  $\text{SiO}_2\text{-Bi}_2\text{O}_3\text{-TiO}_2$  System towards Relative Humidity, *Journal of Chemical Technology and Metallurgy* 53 (6), 1073-1080, 2018

- The characteristics for reading the relative humidity are determined (RH) of sensors in the system  $\text{SiO}_2\text{-Bi}_2\text{O}_3\text{-TiO}_2$ . The samples were synthesized by the method of melt, cooling and controlled crystallization of glass. The sensors were obtained by screen printing of the prepared compositions on aluminum substrates with Pt electrodes. The sensors have been tested in the range (0.0 - 96%) of relative humidity at room temperature. Most of the obtained glass-ceramic materials have a lower sensitivity to RH. The composition  $20\text{SiO}_2.30\text{Bi}_2\text{O}_3.50\text{TiO}_2$  has the highest value of dielectric constant (3000) and the lowest value of dielectric loss (0.0002) at room temperature, which makes it a promising candidate for some industrial applications such as energy storage, electronic capacitors and memory devices.

**[I.3.]** S Slavov, Z Jiao, GLASS-CRYSTALL MATERIALS CONTAINING  $\text{Bi}_{12}\text{TiO}_{20}$  AND  $\text{Bi}_4\text{Ti}_3\text{O}_{12}$  PHASES OBTAINED FROM FREELYCOOLED MELTS OF  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2\text{-Nd}_2\text{O}_3$  SYSTEM, *Journal of Chemical Technology & Metallurgy* 53 (4), 759-764, 2018

- Synthesis of selected samples from the system  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2\text{-Nd}_2\text{O}_3$  is made on the basis of freely cooled melts in the range of 1450 °C to 1100 °C. The initial control of the amount of starting compositions ensures the formation of polyphase glass-ceramics containing  $\text{Bi}_{12}\text{TiO}_{20}$  and  $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ . Amorphous phases are based on oxides of silicon, bismuth, titanium and aluminum: Si-O-Si, Si-O-Ti, Si-O-Al bridge connections and  $\text{SiO}_2$  depolymerized groups and isolated  $\text{TiO}_4$  and  $\text{TiO}_6$  groups. The present study shows a way to control the formation of crystalline and amorphous phases in bulk polycrystalline materials based on a selected system.

**[I.4.]** S Slavov, Z Jiao,  $\text{Bi}_{12}\text{TiO}_{20}$  crystallization in a  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2\text{-Nd}_2\text{O}_3$  system, *Journal of Physics: Conference Series* 992 (1), 012040, 2018

- Polycrystalline single-phase bismuth titanate is obtained by free cooling from a melt heated to 1170 °C. The control of the starting compositions in the system  $\text{Bi}_2\text{O}_3/\text{TiO}_2/\text{SiO}_2/\text{Nd}_2\text{O}_3$  as well as

the thermal gradient of the thermal regimes of heating and cooling leads to the formation of specific structures and microstructures of monophasic silylenite ceramics. The main phase  $\text{Bi}_{12}\text{TiO}_{20}$  incorporated into amorphous network groups based on silicon oxides, bismuth and titanium.

**[I.5.]** Sv Ganev, S Parvanov, S Slavov, A Bachvarova-Nedelcheva, R Iordanova, Y Dimitriev, Influence of  $\text{TiO}_2$  on the thermal stability and crystallization of glasses within  $\text{TeO}_2\text{-Bi}_2\text{O}_3\text{-Nb}_2\text{O}_5\text{-ZnO}$  system, *Bul. Chem. Comm.*, 49, (2017) 103-109

- In this study of the glass in the system  $\text{TeO}_2 - \text{Bi}_2\text{O}_3 - \text{Nb}_2\text{O}_5 - \text{ZnO}$  glasses based on  $\text{TeO}_2$ , containing  $\text{Nb}_2\text{O}_5$  and  $\text{Bi}_2\text{O}_3$  up to 10 mol%,  $\text{ZnO}$  from 5 up to 10 mol%, while  $\text{TiO}_2$  varies from 5 to 50 mol%. The resulting glasses are transparent and yellow ( $\text{TiO}_2$  up to 20 mol%). The thermal stability of the samples was determined by DTA using the difference  $\Delta T$  between the exothermic peak of crystallization ( $T_x$ ) and glass formation temperature  $T_g$  ( $\Delta T = 50\text{--}115^\circ\text{C}$ ). Several crystal phases were identified by XRD, basically  $\text{ZnTeO}_3$  and  $\text{TiTe}_3\text{O}_8$  (in compositions over 20 mol%  $\text{TiO}_2$ ) due to their good dielectric properties. Spectrum analysis shows that the glass network consists mainly of units  $\text{TeO}_4$ . Preliminary dielectric measurements show that the samples have low conductivity and no significant change in dielectric losses up to  $600^\circ\text{C}$ .

**[I.6.]** R Raykov, A Staneva, Y Dimitriev, E Kashchieva, S Slavov, B Blagoev, Glass and glass-ceramics in the  $\text{La}_2\text{O}_3\text{-Gd}_2\text{O}_3\text{-PbO-MnO-B}_2\text{O}_3$  system, *Physics and Chemistry of Glasses-European Journal of Glass Science and Technology Part B*, 56, 4, (2015) 145-148

- The studied compositions in the system  $\text{La}_2\text{O}_3 - \text{Gd}_2\text{O}_3 - \text{PbO} - \text{MnO} - \text{B}_2\text{O}_3$  are located in a section of the phase diagram with a constant content of  $\text{B}_2\text{O}_3$  (30 mol%). For nominal composition  $30\text{B}_2\text{O}_3.8.75\text{La}_2\text{O}_3.8.75\text{Gd}_2\text{O}_3.23.33\text{PbO}.29.17\text{MnO}$ , which is outside in the field of glass formation, a glass-ceramic material is obtained directly from the supercooled melt with a basic crystalline phase rare-earth manganite.

**[I.7.]** S. Slavov, E. Kashchieva, S. Parvanov, Y. Dimitriev, "The effect of Nd substitution of the glass forming ability and thermal properties in the system  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$ ", *Journal of Chemical Technology & Metallurgy*, 50, 4, (2015) 435-440

- The processes for glass formation in the system are studied  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$  wider concentration range. Increasing the content of  $\text{SiO}_2$  leads to a shift of the absorption limit to lower wavelengths. The results of the experiment show that it is possible to obtain low-melting glasses containing 5 mol % - 20 mol %  $\text{SiO}_2$  and 30 mol % - 63 mol %  $\text{Bi}_2\text{O}_3$  by the method of melting and quenching. The replacement of  $\text{Bi}_2\text{O}_3$  with  $\text{Nd}_2\text{O}_3$  up to 10 mol% leads to the production of complex multicomponent glasses and glass-crystal materials with high thermal stability.

**[I.8.]** Anna Staneva, Boris Martinov, Stanislav Slavov, Daniela Karashanova, Janna Mateeva, BMG Melo, Luis C Costa, DIELECTRIC PROPERTIES OF NEW COMPOSITES BASED ON GRAPHENE OXIDE AND NANO-SIZED  $\text{ZnO}$ , *Journal of Chemical Technology & Metallurgy*, 56 (1), 54-66, 2021

- A complete series of graphene oxide and nanosized zinc oxide was obtained and investigated, and the structural and phase composition of the obtained composites was determined. The complex dielectric constant for all samples was measured as a function of frequency. All dielectric characteristics were studied (frequencies from 100 Hz to 1 MHz and temperatures from 150 to 400 K). In the studied temperature range of about 150 K to 400 K and a frequency range of  $10^2$  Hz to  $10^6$  Hz no relaxation processes are observed, and the studied composites have a behavior similar to that of dielectrics with crystal structure and ionic relaxation polarization, which causes increased dielectric losses.

**[I.9.]** P.V. Angelov, S.S. Slavov, Sv. R. Ganev, Y.B. Dimitriev, J.G. Katarov, Direct ultrasonic synthesis of classical high temperature ceramic phases at ambient conditions by innovative method, *Bul. Chem. Comm.*, 45, Special issue A (146 – 152) 2013

- A new approach was used for the synthesis of ferroelectric ceramic materials from the system  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2\text{-Nd}_2\text{O}_3$  based on treatment of precursors with a high-power ultrasonic field generated by a specialized device made to order. When applying the method of synthesis, the high-temperature phase  $\text{Bi}_{12}\text{TiO}_{20}$  was synthesized at room temperature. The formation of this phase is accompanied by the appearance of a small amount of the phase  $\text{Bi}_4\text{Si}_3\text{O}_{12}$  (for samples with the presence of  $\text{SiO}_2$  in batch composition) и  $\text{Nd}_2\text{O}_3$  (for samples with  $\text{Nd}_2\text{O}_3$  presence in the initial composition). Subsequent thermal treatment of these samples (in the temperature range  $500^\circ\text{C}$  -  $1200^\circ\text{C}$ ) shows the formation of the phase  $\text{Bi}_4\text{Ti}_3\text{O}_{12}$  and phase  $\text{Bi}_{12}\text{TiO}_{20}$ .

**[I.10.]** Stanislav S. Slavov, Milena Z. Krapchanska, Elena P. Kashchieva, Yanko B. Dimitriev, "Electrical characteristics of bismuth titanate ceramics containing  $\text{SiO}_2$  and  $\text{Nd}_2\text{O}_3$ ", *Processing and Application of Ceramics* 4 [1] (2010) 39–43

- Bismuth titanate ceramics containing  $\text{SiO}_2$  and  $\text{Nd}_2\text{O}_3$  as additives, is synthesized in two different ways of cooling the melts. The introduction of  $\text{SiO}_2$  and  $\text{Nd}_2\text{O}_3$  leads to more complex crystallization involving several phases, including  $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ . The applied synthesis methods have been shown to be suitable for generating various microstructures in bulk ceramics from bismuth titanate, which is a promising basis for changing their electrical properties. Increasing the content of  $\text{SiO}_2$  improves glass formation, and the addition of  $\text{Nd}_2\text{O}_3$  stimulates crystallization. The conductivity of the selected samples is determined by an impedance analyzer in the frequency range from 10 to 100 kHz and DC resistive bridge using two terminal method. All tested samples are conductive dielectrics  $10^{-6}\text{-}10^{-9} (\Omega \cdot \text{cm})^{-1}$ .

**[II.1.]** SS Slavov, S Soreto Teixeira, MPF Graça, LC Costa, V Popova, Y. Dimitriev,  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-Nd}_2\text{O}_3$  lead-free material for microwave device applications, *International Journal of Applied Glass Science* 10 (2), 202-207, 2018

- Lead-free  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-Nd}_2\text{O}_3$  system and its potential application for microwave devices applications were studied. The samples were synthesized using the melt quenching method. According to the X-Ray Diffraction analysis, the samples are polycrystalline with three different crystal phases. Measurements of the dielectric complex permittivity function were made at 2.7 GHz, using a resonant cavity and the small perturbation theory. The values of the dielectric permittivity of the prepared materials were compared with the commercial ones, and we conclude that these new lead-free materials have a good potential to replace lead-containing devices for microwave frequency applications. The use of polyphase samples in the system  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-Nd}_2\text{O}_3$  may be useful for electronic application at microwave frequencies. The dielectric properties of these investigated samples are interesting, as they present dielectric constants close to the commercial ones (about 7), with low losses (in the order of 10 ). They have the advantage of being lead-free and easy to sinter. The control of the percentage of the observed phases is a challenge to obtain better dielectric properties and their combined influence may be a solution to the problem of the content of harmful raw elements in electronic components.

**[II.2.]** S. Slavov, Y. Dimitriev, GLASS FORMATION IN THE SYSTEM  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$ , *Journal of Chemical Technology & Metallurgy*, 51, 5, (2016) 536-546

- In this study, glass and glass-crystal materials were synthesized in the system  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$ . One limiting condition is that the melting temperature does not exceed  $1450^\circ\text{C}$ , which would limit the losses of components with a lower melting point. The field of glass formation is determined at the cooling rate  $10^2$  K/s. Thermally stable glasses are obtained in the range 10 - 50 mol %  $\text{SiO}_2$ , from 10 to 50 mol %  $\text{Bi}_2\text{O}_3$  and from 10 to 50 mol %  $\text{TiO}_2$  in the central part of the phase diagram. Beyond this limit, glass-crystal materials are involved in the phases:  $\text{Bi}_{12}\text{TiO}_{20}$ ,  $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ . The high content of  $\text{Bi}_2\text{O}_3$  at the expense of  $\text{SiO}_2$  leads to a reduction of the melting point and to lower values of the temperature of the glass phase transition  $T_g$ . One general conclusion is that the glasses are stable up to  $450^\circ\text{C}$  -  $500^\circ\text{C}$ .

**[II.3.]** Stanislav S. Slavov, Elena P. Kashchieva, Svetlin B. Parvanov, Yanko B. Dimitriev „Synthesis of doped bismuth titanate ceramics with  $\text{Nd}_2\text{O}_3$  and  $\text{SiO}_2$  and their electrical properties“ *Journal of Chemical Technology and Metallurgy*, 2, 48, (2013) 174-178

- Bismuth titanate ceramics containing  $\text{SiO}_2$  and  $\text{Nd}_2\text{O}_3$  as additives, is synthesized by a method of cooling the melt in the system  $\text{Bi}_2\text{O}_3\text{-TiO}_2\text{-Nd}_2\text{O}_3\text{-SiO}_2$  in the temperature range  $1260^\circ\text{C}$  -  $1500^\circ\text{C}$ . The addition of  $\text{SiO}_2$  and  $\text{Nd}_2\text{O}_3$  allows control of crystallization, glass formation ability, melting temperature and Curie temperature. It was found that all tested samples are dielectric materials with conductivity between  $10^{-9}$  and  $10^{-13} (\Omega \text{ cm})^{-1}$  at room temperature, dielectric constant from 1000 to 3000 and dielectric losses  $\text{tg}\delta$  between 0,0002 and 0,1.

**[II.4.]** S Slavov, T Tsvetkov, PICARD-LINDELOF ITERATIONS AND MULTIPLE SHOOTING METHOD FOR PARAMETER ESTIMATION, *International Journal of Applied Mathematics* 33 (5), 919, 2020

- In this article, we modify the Picard-Lindelöf iteration scheme in order to show an iteration algorithm for parameter estimation of ordinary differential equations. The proposed algorithm inherited the advantages exhibited in the classical algorithms and, moreover, the parameters can be transformed to a form that are convenient and suitable for computation. In the end, a numerical example has also been discussed to highlight the results.

**[II.5.]** Matej BABIČ, Gyula VARGA, Daniel GHICULESCU, Michal JAKUBOWICZ, Stanislav SLAVOV, George SERITAN, Dragan MARINKOVIĆ, A NOVEL APPROACH FOR PATTERN RECOGNITION BY USING GRAPH THEORY AND ITS APPLICATION IN MECHANICAL ENGINEERING, *ACADEMIC JOURNAL OF MANUFACTURING ENGINEERING*, 19, 3, 2021, ISSN 1583-7904

- In this article, we present a novel approach for pattern recognition by using graph theory and its application in mechanical engineering. One of the most important characteristics of the microstructure of a material is the grain size. The analysis of structure is based on statistical methods and on the principle: from the general image of the structure to the particular (individual grains) by averaging. With computer methods, material structure is characterized by analyzing each grain. When using computer software tools it is also possible to analyze the area occupied by a certain structure directly, without the need for approximate calculations, as well as automatically create a report file. The aim of this paper is to present a new methodology; a novel approach for pattern recognition by using graph theory and its application in the analysis of grain size distribution values. These values were obtained from SEM images of robot laser-hardened specimens using estimating methods to calculate the fractal dimension of 3D objects and calculating the density of 3D visibility graphs.

**[II.6.]** W. Wisniewski, S. Slavov, Ch. Rüssel, Y. Dimitriev, Phase Formation, Crystal Orientations and Epitaxy in  $\text{Bi}_2\text{O}_3/\text{TiO}_2/\text{SiO}_2$  ( $\text{Nd}_2\text{O}_3$ ) Glass-Ceramics, *CrystEngComm* 19, 20 (2017) 2775-2785, <https://doi.org/10.1039/C7CE00542C>

- Four glasses in the  $\text{Bi}_2\text{O}_3/\text{TiO}_2/\text{SiO}_2$  and  $\text{Bi}_2\text{O}_3/\text{TiO}_2/\text{SiO}_2/\text{Nd}_2\text{O}_3$  systems are melted and poured into graphite moulds where they show spontaneous crystallization during cooling. The crystal phases  $\alpha\text{-Bi}_2\text{Ti}_4\text{O}_{11}$ ,  $\text{Bi}_{4-x}\text{Nd}_x\text{Ti}_3\text{O}_{12}$ ,  $\text{Bi}_2\text{Si}_{1-y}\text{Ti}_y\text{O}_5$ ,  $\text{TiO}_2$  and probably  $\text{Bi}_2\text{Ti}_2\text{O}_7$  are shown to crystallize using X-ray diffraction (XRD), energy-dispersive X-ray spectroscopy (EDXS) and electron backscatter diffraction (EBSD). A previously unknown miscibility of  $\text{Bi}_2\text{SiO}_5$  and  $\text{Bi}_2\text{TiO}_5$  is indicated. Oriented layers of monoclinic  $\alpha\text{-Bi}_2\text{Ti}_4\text{O}_{11}$ ,  $\text{Bi}_{4-x}\text{Nd}_x\text{Ti}_3\text{O}_{12}$  and  $\text{Bi}_2\text{Si}_{1-y}\text{Ti}_y\text{O}_5$  are observed. A perfectly epitaxial relationship between  $\text{Bi}_{4-x}\text{Nd}_x\text{Ti}_3\text{O}_{12}$  and  $\text{Bi}_2\text{Si}_{1-y}\text{Ti}_y\text{O}_5$  is proven by EBSD. Crystals with the composition  $\text{Bi}_2\text{Si}_{1-y}\text{Ti}_y\text{O}_5$  with  $y = 0.33, 0.5$  and  $0.66$  are detected.

**[III.1.]** S Slavov, Z Jiao, Bismuth-Titanate  $\text{Bi}_2\text{Ti}_2\text{O}_7$  Crystallization in the  $\text{Bi}_2\text{O}_3/\text{TiO}_2/\text{SiO}_2/\text{Nd}_2\text{O}_3$  System, *Advanced Nanotechnologies for Detection and Defence against CBRN Agents*, 367-372, 2018

- The study demonstrates a way to synthesize compositions from the system  $\text{Bi}_2\text{O}_3/\text{TiO}_2/\text{SiO}_2/\text{Nd}_2\text{O}_3$ , in which only one crystalline phase is present: bismuth titanate pyrochlore ( $\text{Bi}_2\text{Ti}_2\text{O}_7$ ). The synthesis is carried out in two successive stages: initial homogenization of the oxide for 15 minutes and melting at temperatures respectively 1100 °C and 1450 °C. Free cooling to room temperature is done at a rate of about 100 K/min. Using infrared spectroscopy (FTIR) identified the structure and microcrystals distributed in the matrix. Thus, by controlling the starting oxides and the temperature regimes of the synthesis, a single-phase polycrystalline glass-ceramic and a phase-containing ceramic are obtained  $\text{Bi}_2\text{Ti}_2\text{O}_7$ , polymerized  $\text{SiO}_4$  octahedra, featuring  $\text{Bi}_2\text{O}_3$ , as a non-traditional glass former.