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Citation style: Adamczyk Małgorzata, Pilch Michał, Pawełczyk Marian. (2015). Influence of Thermal Treatment on Relaxor Properties of $BaBi_2Nb_2O_9$ Ceramics. "Archives of Metallurgy and Materials" (2015, iss. 2, s. 545-550), doi 10.1515/amm-2015-0012

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A R C H I V E S O F M E T A L L U R G Y A N D M A T E R I A L S
Volume 60 2015 Issue 2
DOI: 10.1515/amm-2015-0012

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INFLUENCE OF THERMAL TREATMENT ON RELAXOR PROPERTIES OF $BaBi_2Nb_2O_9$ CERAMICS

WPŁYW OBRÓBKI CIEPLNEJ NA WŁAŚCIWOŚCI RELAKSOROWE CERAMIKI $BaBi_2Nb_2O_9$

In the hereby paper the implications of thermal modification of $BaBi_2Nb_2O_9$ ceramics in high vacuum (5×10^{-11} bar) at a temperature equal of 1173K are widely discussed. The mentioned modification caused changes in the ions concentration (confirmed by EDS and XPS analysis) and as a consequence an influence on the value of the unit cell parameters as well as on the dielectric and relaxor properties of described ceramics. The obtained results of EDS and XPS analysis of the sample before and after thermal treatment revealed bismuth ions diffusion to the surface of the sample (which was expected) and an almost complete the lack of barium ions on the modified surface. The comparison of EDS and XPS analysis results, suggested that they penetrated the interior of the sample and embedded into the crystal structure in place of bismuth. The hypothesis is in good agreement with the results of X-ray diffraction – the volume of the unit cell had an insignificant increase. The changes in the ions concentration influenced, also in a distinct manner, the dielectric and relaxor properties as well as on the shape of temperature characteristic of thermal stimulated depolarization current observed in BBN ceramics.

Keywords: ceramics, ferroelcectric relaxor, BBN, thermal treatment

W niniejszym artykule są szeroko dyskutowane konsekwencje obróbki termicznej ceramiki $BaBi_2Nb_2O_9$ w warunkach wysokiej próżni (5×10^{-11} bar) w temperaturze 1173K. Wyniki analizy XPS wykonane zarówno przed jak i po obróbce cieplnej ujawniły dyfuzję jonów bizmutu ku powierzchni próbki (co było oczekiwane) i prawie całkowity brak jonów baru na modyfikowanej powierzchni. Porównanie wyników analizy EDS i XPS sugeruje, że jony baru dyfundują do wnętrza próbki i najprawdopodobniej wbudowują się w strukturę krystaliczną w miejsca bizmutu. Ta hipoteza pozostaje w dobrej zgodności z wynikami badań rentgenowskich – objętość komórki elementarnej. Zmiany w koncentracji jonów wpływają również na właściwości dielektryczne oraz zachowania charakterystyczne dla ferroelektrycznych relaksorów, a także kształt charakterystyk temperaturowych prądów termicznie stymulowanej depolaryzacji (TSDC) obserwowanych w ceramice BBN.

1. Introduction

Although the Aurivillius family was already discovered at the beginning of the 1940s [1], these materials were intensively investigated mainly during the last decade. The motivation for this study comes from its excellent polarization fatigue-free behavior and low leakage currents observed in several members of the family (especially $SrBi_2Ta_2O_9$ and $SrBi_2Nb_2O_9$)[2,3]. On the other hand the relaxor behaviour observed in the case of another representative of the family – $BaBi_2Nb_2O_9$ (BBN) is also very interesting not only from the point of view of potential application [4,5], but first of all from the point of view of basic research. Investigations of BBN ceramics were initialized by Smolenski et al [6] and Subbarao [7] in the beginning of 1960s. They reported a considerable diffuseness of the phase transition in the vicinity of 200°C and suggested that it is a consequence of a local disorder in the crystal lattice. At the beginning of XXI century Miranda et al. [8] and A.L.Kholkin et al. [9] initiated investigations of the relaxor properties of the discussed materials. The studies were continued by authors of the following papers [10-15]. Although the properties were a subject of wide discussion their origin is not entirely clear. Shimakawa et al. [12] attributed the relaxor behaviour of BBN to the presence of microscopic clusters with orthorhombic distortion in the macroscopic tetragonal symmetry and the observed ferroelectric properties are a consequence of polarization fluctuations within the (ab) plane induced by these domains. What is more, the structural investigations of $ABi_2Nb_2O_9$ (A=Ba, Sr, Ca) carried out by Blake et al. [13] revealed antisite defects of Bi^{+3} and A^{2+} cations, which the concentration increased with the increasing A cation size. Simultaneously, the orthorhombic distortion decreased. These two various attitudes were not mutually exclusive. Kamba et al. [11] based on the high temperature THz spectroscopy measurements proposed the following scenario: the positional static disorder of Bi and Ba cations was responsible for the ferroelectric relaxor behaviour through the

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formation of nanoregions with different structural distortion levels.

In light of this discussion the paper of Ismunadar and Kennedy seemed to be very interesting [16]. Namely the authors investigated the effect of thermal annealing on the structures of $ABi_2Nb_2O_9$ (A=Ba, Sr) ceramics. The idea of their research was as follows: with increasing temperature the cell volume expanded and this favors the incorporation of the large Ba cations into Bi_2O_2 layers. This arrangement of cations partially remained in their starting placement. However, the quantity of returning cations depended on the manner of quenching (especially depending on the speed of this process). The investigations showed that even 25% of Ba cations could occupy the Bi sites – which was connected with the changes in cell parameters, and should trigger changes of the other properties of ceramics, especially the relaxor behavior.

Therefore the question is what will happen when the amount of bismuth in the interior of the sample changes drastically. In order to answer this question we decided to change the concentration of Bi ions by thermal treatment of the sample in an ultra high vacuum (UHV). Taking into consideration the value of the sublimation's temperature of pure bismuth, which was equal to 1023K, and taking into account the possibility of increasing this value for Bi connected to the crystal lattice, we decided to carry out the mentioned thermal treatment at a temperature equal to 1173K. The drastic changes in bismuth concentration were expected. So attempts to investigate the chemical state of sample's surface before and after thermal treatment in a UHV by using the XPS measurements were made. According to the thesis of other authors presented above we expected an improvement in the behavior characteristics for ferroelectric relaxors.

2. Ceramics preparation

The $BaBi_2Nb_2O_9$ ceramics were prepared using the conventional mixed-oxide processing technique. Stoichiometric amounts of $BaCO_3$, Bi_2O_3, Nb_2O_5 reagents were weighed and mixed. Thermal synthesis of the pressed mixture was carried out at 1223K for 2h. Then the crushed, milled and sieved materials were pressed again into cylindrical pellets and sintered at 1373K for 6h and then cooled to room temperature for 12h. The obtained ceramics were semitransparent and of a good mechanical quality. All the samples, were cut using diamond saw, polished in order to obtain flat and parallel surfaces. The samples for dielectric and thermal stimulated depolarization current measurements were coated with silver electrodes, using an appropriate silver paste (without thermal treatment). After measurements the electrodes were removed and the surfaces were very precisely cleaned. Next samples were thermally treated in an UHV (5×10^{-11}bar) at the constant temperature 1173K for one hour and cooled down to room temperature. The samples used for dielectric and TSDC measurements were again coated with silver paste.

The grain structure and the distribution of all elements throughout the grains were examined before thermal treatment by the scanning electron microscope (SEM), JSM – 5410, with an energy dispersion X-ray spectrometer (EDS) by Oxford Instruments.

The X-Ray Photoelectron Spectroscopy measurements were conducted on ceramic samples, before and after the thermal treatment. The test was conducted using the Physical Electronics Spectrometer PHI 5700 equipped with Perkin Elmer instruments using Al Kα monochromatic X-ray source with energy of 1486.6 eV, in ultrahigh vacuum (UHV) conditions, from the area with 0.8 mm radius. The energy resolution was 0.3eV. All photoelectron spectra were calibrated against the peaks of Au $4f_{7/2}$ at 83.98 eV, Ag $3d_{5/2}$ at 368.27 eV and Cu $2p_{3/2}$ at 932.67 eV of binding energy. The test of the surfaces of the ceramics was carried out a takeoff angle of 45°. The electron float gun was used for the compensation of positive surface charge, which may appear on the insulator materials surface. The XPS measurement was carried out for the core lines of Bi 4f, Ba 3d, Nb 3d, O 1s, C 1s, and valence band. However we discussed only the survey lines. All measurements were performed under UHV conditions in the order of 5×10^{-11}bar at 1173K and 5×10^{-13}bar at room temperature. The angle between the X-Ray source and the sample surface was 45°. The Multipak Physical Electronics program enabled quantification of the XPS spectra utilizing a peak area and peak height sensitivity factor. The standard atomic concentration calculation provided a ration of each component to the sum of the other elements taken into account in the data. Only those lines clearly visible in the spectrum were considered. For those survey lines the background was not subtracted, the limit of the region of the line was individually selected, and after integration was done [17].

XRD measurements were carried out using the Huber diffractometer (Seemann – Bohlin geometry) with monochromatic Cu-Kα_1 radiation (30kV, 30mA). The angle scale of the received diffraction diagrams was scaled to 2θ (Bragg – Brentano geometry) by the Au standard (JCPDS number 12-0403). The diagram was measured from 20 to 100° in 2θ with 0.05° steps. The calculations were performed using a package of programs for treatment of powder X-ray diffraction data DHN-PDS. In order to receive the the exact location, intensities and widths of diffraction lines, the experimental data was fit using theoretical functions. The attempts of fitting by Gaussian (G), Lorenzian (L), modified Lorenzian (ML) and intermediate Lorenzian (IL) were made. The IL profile showed the best agreement with experimental data and this profile was taken to fit all segments of the diagram including diffraction lines. The data received in this manner was used to calculate the unit cell parameter.

The temperature dependence of the dielectric response of the BBN ceramics was investigated from 300 K to 720K using the impedance analyzer HP 4192A.

Additionally the measurements of thermally stimulated depolarization currents were made. Samples before and after thermal treatment in an UHV first polarized in a DC field with a strength E_p equal to 1kV/cm applied for 10 minutes at a temperature T_p =473 K and cooled in the field to 323K where the field was switched off. Next the samples were heated with a constant rate of 5K/min to the temperature of 723K.

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3. Results and discussion

3.1. EDS microanalysis

Before thermal treatment the microstructure of samples was investigated by using a scanning electron microscope. The average size of the grains was equal to 2 μm. The measurement was repeated after thermal treatment, which allowed the conclusion that microstructure did not reveal any changes. The same conclusions were drawn for 0.86PZN-0.05BT-0.009PT (PZN-BT-PT) ceramics annealed in air [18]. Quantitative microanalysis was performed with the ISIS – 300 SEMQuant program for individual grains of the sample before and after thermal treatment and all elements were normalized to 100%. The results are collected in the Table I.

TABLE 1
Variation of barium, bismuth and niobium obtained by using the EDS analysis

	Bi [%]	Ba [%]	Nb [%]
Nominal content	40	20	40
As sintering sample	39.48	20.63	39.89
Annealed sample	27.78	12.79	59.43

The stoichiometry of the sample before thermal treatment is shown to agree with the nominal content of elements. After treatment the deficiency of barium and bismuth ions was observed. However, the results of the EDS analysis was not sufficient to determine the direction of diffusion, what led the authors to make a decision about the XPS measurements in order to test the surface of thermal treatment sample.

3.2. XPS measurements

The XPS spectrum of the $BaBi_2Nb_2O_9$ ceramics was measured at room temperature twice before and after the thermal treatment, respectively. The results are shown in Fig. 1a and 1b. Any contamination, besides a vestigial amount of carbon and lead, was not found in the spectrum. The impurities originated from the applied technology processes. These spectra were used for the atomic concentration calculations using the Multipak program. The results of the calculation of concentration for each element for the as-sintered sample are comparable with the EDS results. Deviations from the stoichiometry are observed for the thermally modified sample. The chemical compositions of the thermally treated sample were obtained as: 54.1 at. % for Bi, 0.3 at. % for Ba, 45.6 at. % for Nb.

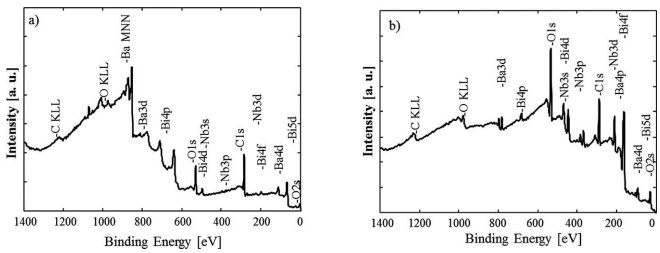

Fig. 1. The XPS spectrum measured in wide energy range of the $BaBi_2Nb_2O_9$ ceramics a) before and b) after thermal treatment at 1173K in the ultra high vacuum

The results were surprising and pointed to the processes taking place on the sample during thermal treatment to be more complicated than initially assumed. The initial scenario assumed pure bismuth sublimation. However the XPS measurements revealed two competitive processes. Namely thermal treatment in the reducing atmosphere caused a reduction of Bi_2O_3 into Bi metal [19], was as expected. The bismuth ions diffused from the interior of the sample to the surface and remained there. Simultaneously the barium ions "disappeared" from the surface. Hypothetically, it could be assumed that barium ions diffused into the sample and took the place of the bismuth gaps. The results of investigations carried out by Ismunadar and Kennedy [16] revealed that part of barium ions wrongly built in the crystal lattice strongly depended on the history of thermal treatment. The high temperature caused expansion of the crystal structure and favored the incorporation of the larger Ba cations into the Bi_2O_2 layers in place of bismuth vacancies or in the internodal position.

Such a scenario is connected with the creation of new point defects. Namely taking into consideration the results of investigations of defects chemistry of SBN ceramics proposed by A.C.Palanduz [20-22], it can be assumed that the simple exchange of barium and bismuth cations resulted in the presence of Ba'_{Bi} acceptor centers in Bi-layer, and $Bi^{\bullet}_{Ba}$ donor centers in the perovskite – like layers. In an isotropic structure such place exchange should be self-compensating, and no other defects would be required for charge balance. But in the case of a layered structure the self-compensation process was not fully effective and there was a significant amount of charge compensation within each layer. Thus Ba'_{Bi} acceptor centers in the Bi layer can be partially compensated by oxygen vacancies, $V^{\bullet\bullet}_{O}$ or holes, $h^{\bullet}$, while the $Bi^{\bullet}_{Ba}$ donor centers in the perovskite layers can be partially compensated by cations, vacancies V_{Ba}" or electrons e'. The diffusion of bismuth ions up to the surface and barium ions into the interior of the sample intensified the process of defect creation. Due to large differences in the size of the ionic radius of Ba and Bi ions outlined in the above scenario both should be accompanied by: large lattice strain and increase of unit cell volume (as suggested by the authors of paper [16]).

3.3. XRD measurements

The XRD measurements were done with the aim to confirm the hypothesis concerned in the processes occurring on a sample during the thermal treatment in a reducing atmosphere. The X-ray diffraction pattern was obtained for the sample before and after thermal treatment (Fig. 2).

The results are shown to be in a good agreement with the JCPDS standard number 12-0403 for the $BaBi_2Nb_2O_9$. All of the line indexes related to the Aurivillius structure were assigned. In case of the thermal treatment sample the obtained diffraction peaks were significantly more broadened, which indicated the presence of large strains of a crystal structure, as suggested earlier. The lattice parameter obtained from the X-ray pattern for non-thermal modified sample was in a good agreement with the results reported in paper [16]. After thermal treatment the value of the parameters increased (see Table II) and the volume of the crystal unit cell changed about 4.4%. The XRD results were an indirect confirmation of the assump-

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tions concerning the diffusion of barium ions to the interior of the sample and their incorrect build into the crystal lattice.

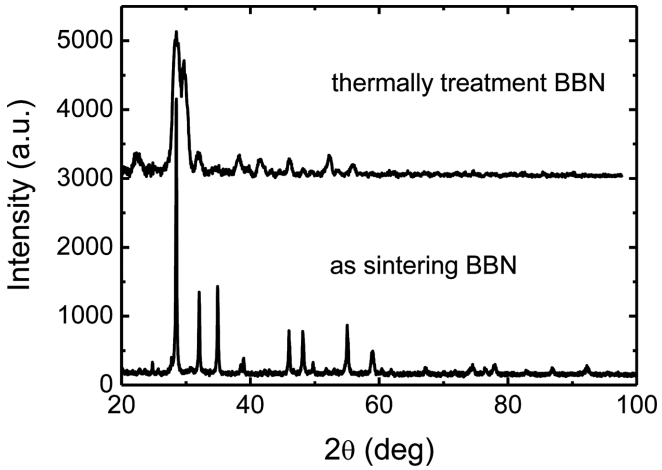

Fig. 2. X-ray diffraction pattern of $BaBi_2Nb_2O_9$ ceramics before and after thermal treatment at 1173K in the ultra high vacuum

TABLE 2
The value of lattice parameter before and after thermal treatment

Sample	a=b [Å]	c [Å]	V[Å^3]
Before thermal treatment	3.9406±0.0006	25.6378±0.0059	396.11
After thermal treatment	3.9862±0.0091	26.1495±0.0794	415.51

3.4. Dielectric and thermally stimulated depolarization currents measurements

The comparison of the temperature characteristic of real part of dielectric permittivity ε‘, obtained at the measuring field of a frequency of 1kHz for the samples before and after annealing in an ultrahigh vacuum is given in Fig. 3.

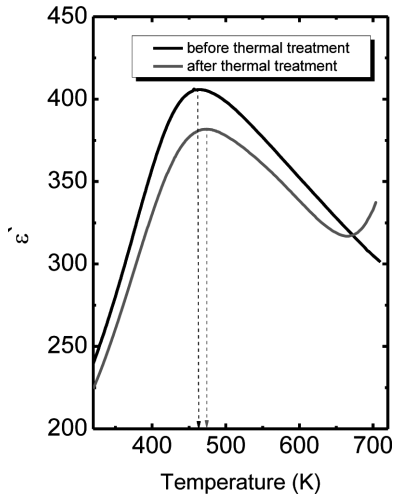

Fig. 3. Real part of permittivity as a function of temperature measured on heating at frequency of measuring field 100 kHz for sample of BBN ceramics before and after thermal treatment at 1173K in the ultra high vacuum

The shape of these dependencies pointed at the strong diffuse character of phase transition for both non-thermal treatment and thermal treatment samples. The quantitative assessment of the diffusivity was possible due to the formula proposed by Martinera and Burfoot (1)[23]

$$\frac{1}{\varepsilon'} - \frac{1}{\varepsilon'_{\max}} = \frac{C}{(T - T_m)^{\gamma}} \tag{1}$$

where ε'_{max} is the maximum value of the real part of the dielectric permittivity at the temperature (T_m), C is the Curie-like constant and γ is the degree of diffuseness. The limiting values of 1 and 2 for γ, respectively, reduced the expression to the Curie-Weiss law valid for a normal and the quadratic dependence valid for the ideal relaxor ferroelectric. The thermal treatment changed the value of γ from γ =1.45±0.02 to γ =1.68±0.02. The increase in the value of γ implied that there was an increase in the diffuseness of the transition, which was connected with the growing ions disorder.

Moreover the thermal treatment influence also on the value of the dielectric permittivity, caused its decrease. The results were in a good agreement with the results presented by A.Z. Simoes et al [24]. Namely the excess of bismuth caused the increase of the dielectric permittivity value of BBN ceramics, so its deficiency should have an influence in an opposite manner. The static disorder of barium and bismuth cations was also responsible for the property characteristics of the ferroelectric relaxor. In light of the thesis presented above the relaxor behaviour should be more distinct after thermal treatment. Aiming at confirmation of these assumptions we measured the temperature characteristic of dielectric permittivity at various frequencies, and the results are given in figure (Fig. 4).

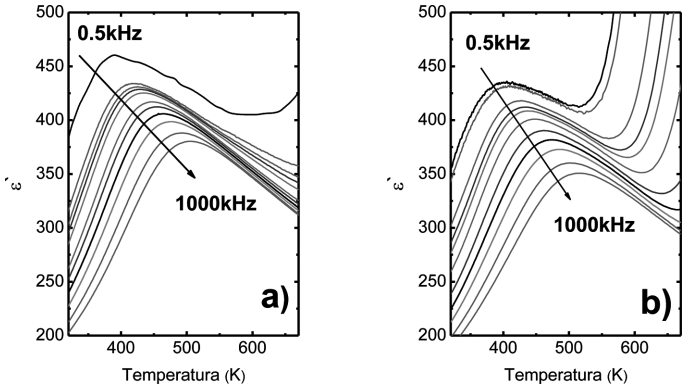

Fig. 4. Real part of permittivity as a function of temperature and frequency of measuring field 100kHz obtained on heating for $BaBi_2Nb_2O_9$ ceramics a) before and b) after thermal at 1173K in the ultra high vacuum treatment

Both discussed samples have shown a reduction of $\varepsilon'_{\max}$ and a shift of the corresponding temperature T_m towards the higher values with frequency increase. However the behavior was more significant for the thermal modified sample, namely the value of dispersion‘s degree (defined as the difference between T_m measured at 1000 kHz and 0.5 kHz) increased from ΔT_m = 94 K before thermal treatment to the ΔT_m =121 K after the process.

It is commonly known that in relaxor ferroelectric materials the crucial role play the polar nano-regions, which appear at the temperature distinct higher than Tm. The origin of

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mentioned regions is connected with chemical heterogeneity. Simply at mentioned temperature range the microstructure of the ferroelectric relaxors consists of polar nanoregions embedded in a nonpolar matrix. They are mutual independent. Two types of fluctuations are related to the mentioned regions:

– hetero-fluctuations – i.e. the appearance/ disappearance of the dipole moments
– orientation fluctuation associated with the polarization vector reorientation.

The second kind of fluctuation is more probably when the temperature decreases and the number of polar nanoregions and their size increase, these interactions with each other appear. At the freezing temperature fluctuations become infinitely slower and the system enters into a so-called polar glass state.

Within this model the frequency dependence of the dielectric permittivity maximum temperature (T_m) described the well known empirical Vogel – Fulcher relationship (2)

$$f = f_0 \exp\left[\frac{-E_a}{k(T_m - T_f)}\right] \tag{2}$$

where E_a is the activation energy, T_f is the freezing temperature of polarisation fluctuation, and f_o is the pre-exponential factor. This fact allowed us to determine the values of E_a, T_f and f_o for both samples: as sintering and thermal modified. The obtained data is presented in Table III. The significant decreasing of the activation energy value and the shift of the freezing temperature to higher value was observed in the thermal modified sample, which pointed to the crucial role of barium and bismuth ions concentration in determining of ferroelectric relaxor properties in BBN ceramics.

TABLE 3

Freezing temperature (T_f), activation energy (E_a) and pre-exponential factor f_o

Sample	T_f (K)	E_a (eV)	f_o(Hz)
Before thermal treatment	170	0.46	$9.68*10^{12}$
After thermal treatment	303	0.29	$4.2*10^{10}$

The detailed analysis of temperature dependence of dielectric permittivity for non-thermal treated samples revealed the second, very weak, low frequency dispersion at high temperatures of the paraelectric phase (Fig. 4a). This dispersion is a common feature in a lot of different ferroelectrics (particularly single crystals and ceramics characterized by ABO_3 perovskite structure) [25-28] and connected with the space charge present in the sample. A clear link exists between the mentioned dispersion and sample conductivity. The thermal treatment caused an increase of the space charge participation in the bulk of the sample, which led to the increase of both conductivity and dielectric dispersion [27]. Moreover the important role of space charge in screening polar nanoregions should not be forgot. The problem was widely described in papers [29,30-32]. The one of ways of minimizing depolarization field energy associated with spontaneous polarization of nanoregions is to screen them by the space charge of the surrounding space. Even after the disappearance of the P_s inside the screened nanoregions, at a high enough temperature, the trace of non-random distributed space charges remain in the PE matrix because of their long relaxation time. At high enough temperatures the distribution of ion defects is thermally randomized. The liberated way of such electron and ionic space charges, together with the carriers injected from electrodes, caused the observed increase of the thermally stimulated depolarization current (Fig. 5).

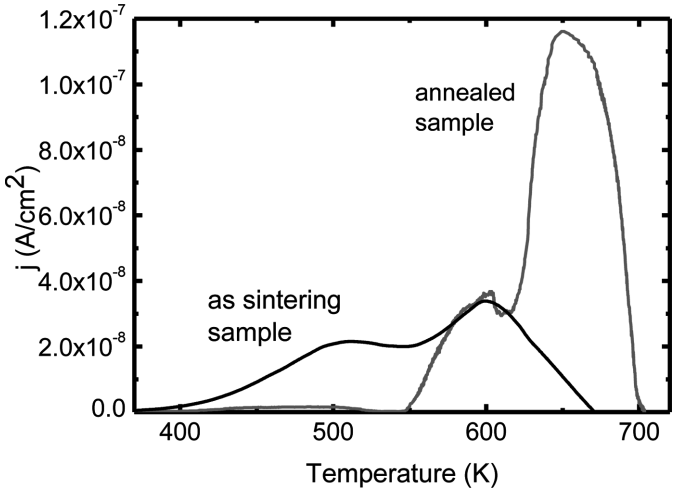

Fig. 5. Thermally stimulated depolarization currents vs. temperature for BBN ceramics before and after thermal treatment at 1173K in the ultra high vacuum

The temperature characteristic of TSDC changed drastically for thermal treated sample (Fig. 5). Namely the first peak stayed at the same level, but at the higher temperature appeared a second larger maximum. In the authors' opinion the second maximum may be associated with the appearance in the bulk of sample additional defects, which created the dipolar defect complexes (for example oxygen and barium vacancies), as is the case for Dy doped $BaTiO_3$ [33]. The complexes contribute to the growth of mentioned low frequency dielectric dispersion and are responsible for the second maximum observed on TSDC characteristic.

4. 4. Conlusions

In the preliminary assumption the thermal treatment of $BaBi_2Nb_2O_9$ ceramics sample was applied in order to reduce the bismuth ions concentration. The aim has been achieved. The XPS and EDS measuremets revealed the changes of concentrations of bismuth, however also revealed were the changes in barium concentration. The thorough analysis of EDS and XPS results as well as XRD measurements led us to the formation of the following scenario: the bismuth ions diffused to the surface of sample and remained there, whereas the barium ions moved in the opposite direction and penetrated the interior of the sample taking the place of the bismuth gaps. As a consequence the value of the volume of the crystal cell increased and the large strains of the crystal structure were presented in the ceramics.

The described above results of the dielectric measurements clearly indicated that the relaxor properties are inextricably linked with the barium and bismuth concentration. Namely the dispersion of $\varepsilon'_{\max}$ and T_m values, characteristic

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for the ferroelectric relaxor, increase, whereas the activation energy of the flipping mechanism of polarization inside the polar regions decreased. The facts point to that the disturbances in barium and bismuth concentration is determining the ferroelectric relaxor properties. Moreover the mentioned changes of concentration make easier to crossover to the polar glass state.

Acknowledgements

The authors would like to thank Prof. dr hab. M.Sopicka-Lizer for helpful discussion and remarks.

This work was supported by grant N N507504338 from the Ministry of Science and Higher Education in Poland.

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Received: 20 September 2014.

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