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 53 2008 Issue 3

P. RANACHOWSKI*, F. REJMUND*, A. PAWEŁEK**, A. PIĄTKOWSKI**

# STRUCTURE DEGRADATION, TEXTURE AND ACOUSTIC EMISSION IN COMPRESSED CORUNDUM CERAMICS

# TEKSTURA, DEGRADACJA STRUKTURY ORAZ EMISJA AKUSTYCZNA W CERAMICE KORUNDOWEJ W PROCESIE ŚCISKANIA

The paper presents the results of microscopic, ultrasonic and acoustic emission (AE) investigations of corundum ceramic material C 799 kind samples, exposed to compressive stress. On the basis of AE measurements of slowly compressed samples, the successive stages of structural degradation have been registered. Microscopic analysis enabled to specify the processes of the gradual growth of microcracks and crushing out of corundum grains, similarly to the aging processes occurring in material after long period of exploitation under mechanical stress. The analysis of AE results revealed diversified strength and mechanical-acoustic characteristics of the particular samples. It was found that this effect was due to technological defects, especially textural inaccuracies – different grain size and their non-uniform spatial distribution.

The structure analysis of the samples compressed to advanced subcritical stage revealed the presence of micro- and macrocracks. The most of the cracks underwent propagation along grain boundaries, whereas only few within the grains. It was found that the centers of joined bigger grains are especially susceptible to the destruction process. During the critical stage of destruction the propagation of cracks occurs at great velocity and throughout all the elements of the structure.

Investigations revealed that differences registered for the strength and the mechanical-acoustic characteristics are due to the inhomogeneity of the material in the semi-macro as well as in the micro scale. The occurrence of groupings of grains in the structure of the corundum material represents most probably the intermediate state, leading to the known effect of the abnormal grain growth (AGG).

*Keywords*: corundum material, compressive strength, structural degradation, acoustic emission, ultrasonic testing

W pracy przedstawiono wyniki badań mikroskopowych, ultradźwiękowych oraz emisji akustycznej (EA) próbek poddanych wolno narastającemu obciążeniu ściskającemu. Przedmiotem badań były małogabarytowe kształtki z tworzywa korundowego rodzaju C 799. Zawartość tlenku glinu w badanym materiale wynosiła 99,7%. Z uwagi na szerokie zastosowanie we współczesnej technice, badania procesów degradacji tworzyw korundowych mają istotne znaczenie poznawcze. Przeprowadzone badania umożliwiły rozpoznanie tekstury tworzywa oraz monitorowanie kolejnych etapów degradacji struktury, aż do całkowitego jej zniszczenia. Zastosowana metodyka badań pozwala na odniesienie wyników do procesów starzeniowych, zachodzących w trakcie wieloletniej eksploatacji elementów korundowych. Wykonane pomiary wykazały zróżnicowaną wytrzymałość oraz charakterystykę mechaniczno-akustyczną poszczególnych próbek. Obserwacje mikroskopowe struktur w różnych fazach degradacji ujawniły, że zróżnicowane właściwości próbek są konsekwencją defektów technologicznych, wielomodalnego rozkładu wielkości ziarn oraz niejednorodnego ich rozmieszczenia (tekstury materiału). Autorzy uważają, że rejestrowana struktura próbek odpowiada wstępnemu etapowi znanego efektu nadnaturalnego wzrostu ziarn (abnormal grain growth).

## 1. Introduction

Corundum materials with high alumina ($Al_2O_3$) content, exceeding most often 80% – are widely applied in present day technology. They are used first of all as elements of technical devices for which high mechanical, thermo-mechanical strength or abrasive resistance are required. This material is resistant to oxidation, chemical corrosion and various types of irradiation. Its thermal conductivity is similar to that of stainless steel. Moreover, the corundum material has good electric parameters – high dielectric constant and low loss angle. In recent times it is also used for the production of the carrying rods of hybrid insulators [1].

In this study there are presented the mechanical-acoustic and structural investigations of corundum material of C 799 type and $Al_2O_3$ content

* INSTITUTE OF FUNDAMENTAL TECHNOLOGICAL RESEARCH PAS, 00-049 WARSZAWA, 21 ŚWIĘTOKRZYSKA STR., POLAND
** INSTITUTE OF METALLURGY AND MATERIALS SCIENCE, POLISH ACADEMY OF SIENCIES, 30-025 KRAKÓW, 25 REYMONTA STR., POLAND

equal to 99.7% [2-4]. For the fabrication of the samples the granulated product NM 9922 of Nabaltec firm was used. The size of grains collected in the aggregates was than 0.5 $\mu$m smaller. Samples for the investigations were formed using the method of single axial pressing (10 MPa) and condensed isostatically (120 MPa). After firing into biscuit at the temperature 1250 °C, samples intended to be used in the investigations were cut out from a larger element, taking into account grinding and shrinking of the mass. Next, the samples were fired at the maximal temperature 1700 °C and stored for 1.5 hr at the sintering temperature. The samples were ground to obtain the dimensions 5×5×10 mm, then their density was determined and the absorbability as well as imperviousness and absence of cracks in the alcohol solution of fuchsine were controlled. The obtained material had the density $\rho$=3.89 g/cm$^3$ and did not contain any detectable defects. In order to determine the size of grains some samples were polished and as well as thermally etched at the maximum temperature 1300 °C and stored for 1 hr at this temperature.

Ultrasonic control of the homogeneity of the samples revealed a slight anisotropy as well as some differences of the acoustic parameters and the elasticity modulus between the samples. For example, the velocity of the longitudinal waves $c_L$, measured in the direction perpendicular to the axis for various samples amounted from 10 480 to 10 600 m/s (the inaccuracy of measurement was equal to ± 20 m/s). Young's elasticity modulus E determined in the same direction was enclosed, depending on the sample, in the range from 364 to 373 GPa. The average E value was 368 GPa, at the measurement inaccuracy equal to ± 2 GPa. The mean value of Young's modulus in the direction parallel to the sample length was somewhat lower and was equal to 360 GPa. The obtained parameters considerably exceed the required by standard values of $\rho$ = 3.70 g/cm$^3$ and E=300 GPa for the corundum material of C 799 type [4]. The amplitude damping coefficient for longitudinal waves of the frequency f=10 MHz showed also some differences. In the direction perpendicular to the sample axis it was contained in the interval 1.0 ÷ 1.2 dB/cm, whereas in the parallel direction it was 0.9 ÷ 1.1 dB/cm, at the measurement inaccuracy below 0.1 dB/cm. Samples compressed to subcritical level of structure degradation were characterized by nearly unaffected velocities of waves propagation $c_L$, while damping coefficient $\alpha$ increased to high values in the range 2.5 ÷ 3.2 dB/cm.

## 2. Mechanical-acoustic research

The samples were subjected to mechanical-acoustic measurements using the technique of acoustic emission (AE) on a special two-channel measuring system. Pieces of small dimensions were put to slowly increasing compressive stress (v=0.02 mm/min), with simultaneous registration of the force in one channel, and AE descriptors in the second one. The arrangement of the measurement system and the method of investigations were described in detail in the papers [5,6,7]. The research enabled the recording and description of the correlation between the increasing external load and the processes of structure degradation, which are reflected in the activity AE. Acoustic method is suitable for the investigation of the destruction of ceramic materials, due to the fact that initiation and growth of microcracks belong to the main sources of AE signals. Examination of alumino-silicate and corundum ceramic materials enabled to state that the sum of AE events during the loading period is a good descriptor of the intensity of the processes of cracking, which are the cause of mechanical degradation of the material. There exists a correlation between the rate of the increase of cracks and the rate of AE events (number of AE events per unit of time) [8]. Registration of this descriptor allows monitoring the process of destruction of the microstructure of a ceramic material under load. The authors stated as well good correlation between the processes of material structure degradation, mainly connected with microcracks development, and the AE activity represented by the effective value of AE signal (RMS).

There exist serious analogies between the effects of many years long exploitation under load applied to the material and the compressive stresses in a relatively short lasting laboratory test. This observation has been proved by investigations, carried out by the authors, on ceramic aluminous materials. However, it is necessary to apply a quasi-static, very slow increase of stress and a precise registration of the AE descriptors [7,9].

The compressive strength of the samples has shown an unexpectedly wide distribution. The mean strength of ten destroyed specimens was 3 163 MPa. The least resistant of the samples underwent decohesion already at the stress equal to 2 043 MPa. It was found impossible to measure the strength of the most resistant sample. For technical reasons it was necessary to stop the process of loading at the stress of 3 804 MPa. Relative dispersion of strength was equal to over 56 %. The mechanical-aco

-ustic characteristics of the particular samples showed high differentiation – figures 1, 2 and 3. At stresses below 2 500 MPa only some samples, of more inhomogeneous structure, showed single AE signals of various intensities. Among them was the least resistant of the investigated specimens – figure 1. If there appear the intervals of continuous acoustic activity – the AE signals are most frequently characterized by very low energy – figures 2 and 3. Above the stress 2500 MPa the loaded

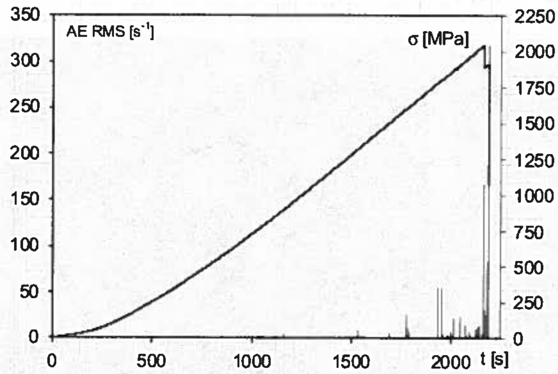


Fig. 1. The course of RMS AE rate versus the increase of compression stress for the least resistant sample of the strength 2 043 MPa

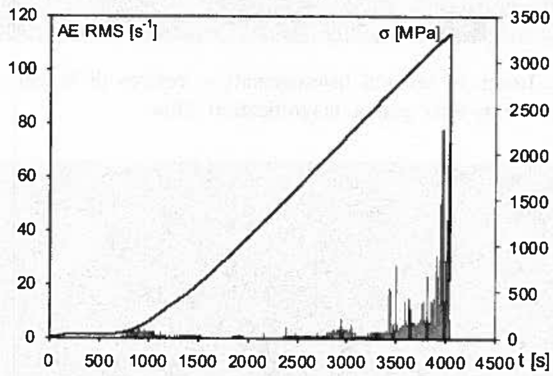


Fig. 2. The course of RMS AE rate versus the increase of compression stress for the sample of the strength 3 320 MPa

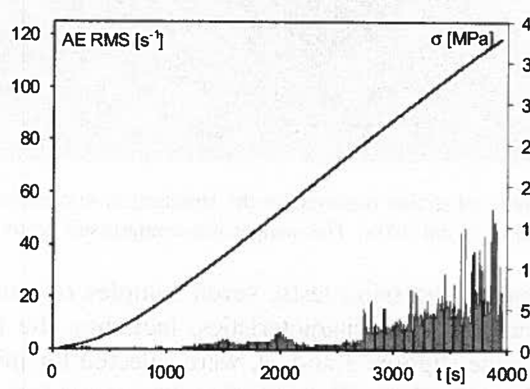


Fig. 3. The course of RMS AE rate versus the increase of compression stress for the most resistant sample. Compression was stopped at 3 804 MPa

samples showed AE effects in the form of single signals, occasionally forming intervals of continuous acoustic activity. The interval of subcritical stresses was characterized by diversified width – depending on the sample strength. The subcritical stage preceded a short critical interval, containing a group of signals of high energy. The critical interval occured in the range of stress between 30 and 50 MPa and it directly precedes the decohesion of the sample. Thus, the stress at which the critical stage occurs is closely connected with the sample strength.

## 3. Structural research

In order to explain differentiation in the values of the destructive stresses for the particular samples and their various mechanical-acoustic characteristics, it was necessary to carry out accurate microscopic investigations of the corundum material structure. The research concerned the size and the uniformity of distribution of the grains and the effects of the compressive stress on various parts of the samples. The special attention was paid to presence of defects in the structure before and after compression of the samples.

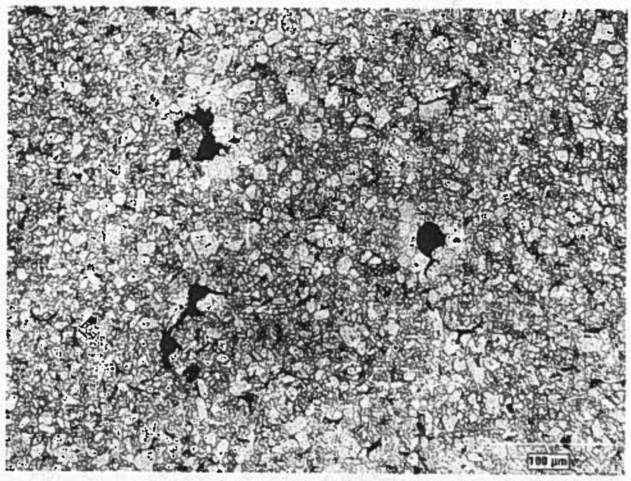


Fig. 4. Technological defects observed in the structure of corundum material in magnification 200×. Large pores and fissures are visible

The majority of samples showed distinct inhomogeneities of the structure in the semi-macro scale as well as insufficient homogeneity in the micro scale. These samples contained fine technological defects such as gaseous and solid inclusions, fissures and partly broken grain boundaries – figures 4 and 5. The great majority of the samples showed a bimodal size distribution of the grains – figure 6. The sizes of bigger grains were most often in the range from less than 10 to over 30 $\mu$m, whereas the diameter of fine grains was from a fraction of micrometer to a few micrometers. Only some samples demonstrated uniform size of grains and proper internal texture – figure 7. For example the grains of the most resistant sample showed one-modal size distribution. Their diameters were enclosed in a narrow range from above 1 to 14 $\mu$m, with the mean value equal to 7 $\mu$m.

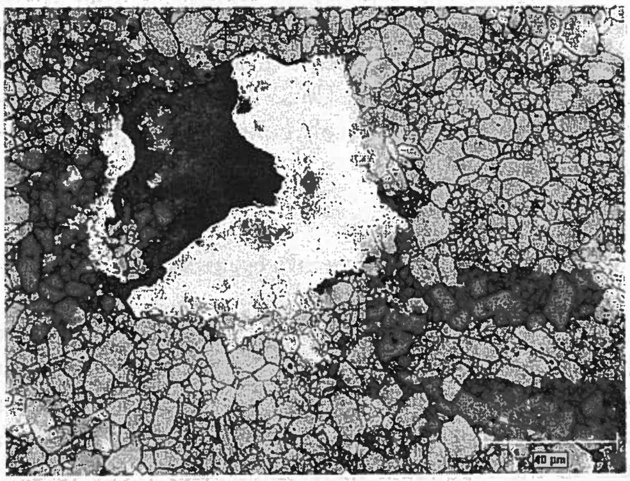

Fig. 5. Large gaseous inclusion in the structure of corundum material, formed probably as a result of burning of plasticizer, magnification 500×

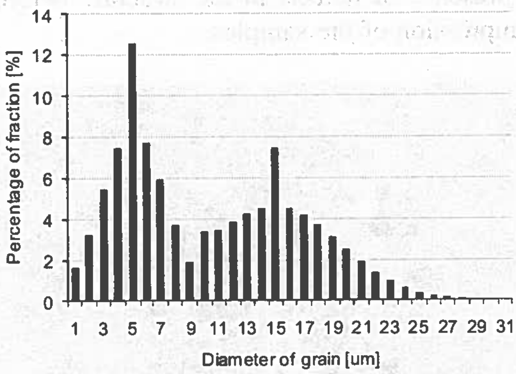


Fig. 6. Typical bimodal distribution of the grains diameters, obtained for the sample of examined corundum material

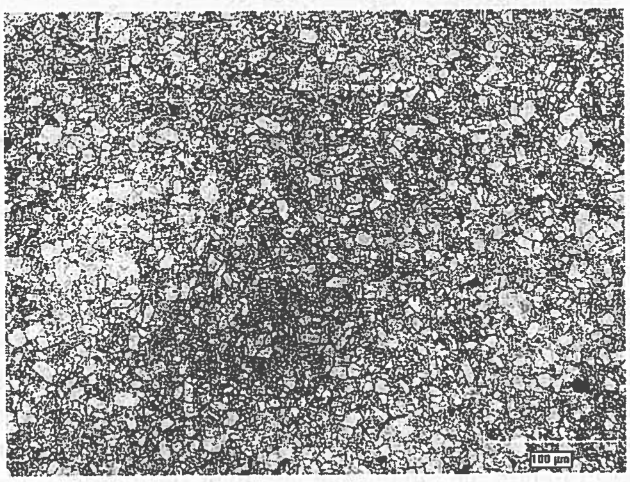

Fig. 7. Proper texture and uniform grains' sizes distribution of the corundum material in magnification 200×. Visible structure loosening is the consequence of compression of the sample up to 3804 MPa

There were observed textural heterogeneities in the structure of the samples. The bigger grains often formed centres of the size of the order of tens micrometers (up to 100 $\mu$m), surrounded by smaller areas containing finer grains – figure 8. Another discovered inhomogeneity was arrangement of the structure in form of bands. The greater and/or the smaller grains were organized as separate bands having width of the order of some tens of micrometers – figure 9. The most regular structure was observed in case of the mentioned sample of the highest strength (figure 7).

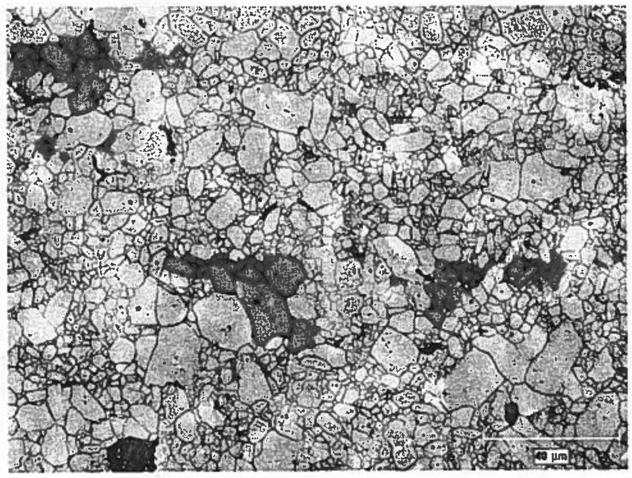

Fig. 8. Image of textural heterogeneity – centres of bigger grains surrounded by finer grains, magnification 500×

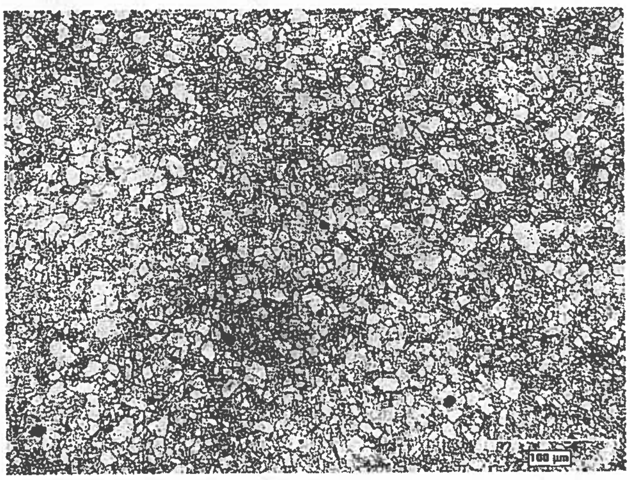

Fig. 9. Bands of grains observed in the structure of corundum material, magnification 200×. The sample was compressed up to 3230 MPa

After compression tests, seven samples of various mechanical-acoustic characteristics, including the most resistant one (figures 3 and 7), were selected for microscopic investigations. Their loading was stopped shortly before the critical stage of structure destruction. In all the samples, there were observed the effects of structure loosening. This concerned especially the central part of the samples where the stresses were cumulated. Propagation of microcracks occurred almost only along the grain boundaries. Nevertheless, there have been observed places of crushed out grains or parts of grains. These areas covered 0.2÷0.3 % at the boundaries and up to 1.0 % of the surface of a polished section in the middle of the samples, whereas initial porosity of the material in any of the samples did not exceed 0.11 %. Figure 10

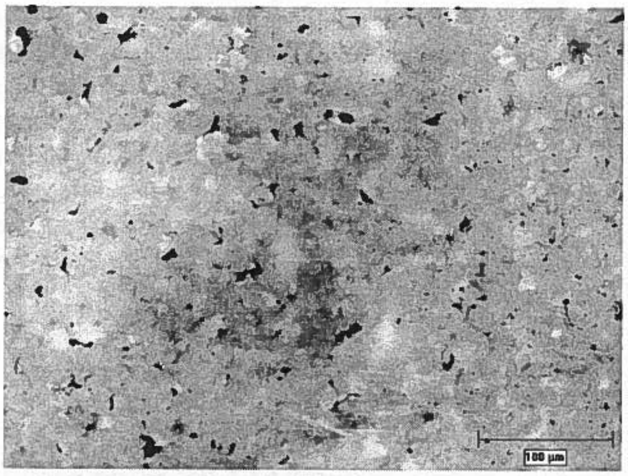


Fig. 10. Image of the structure in the middle part of the sample loaded up to 3 180 MPa, magnification 200×

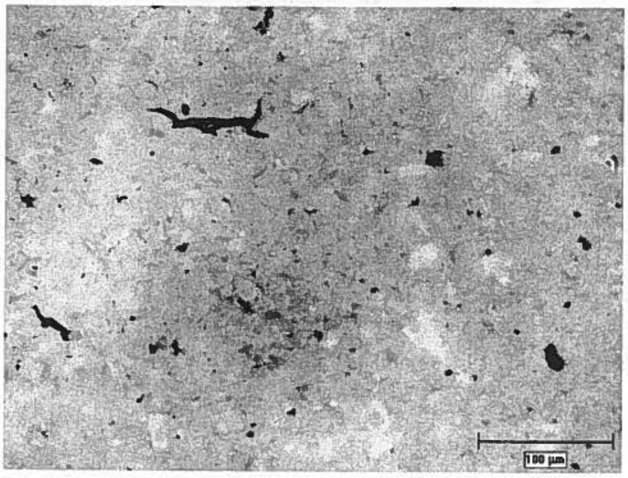


Fig. 11. Structure of the central part of the sample compressed up to 3200 MPa, magnification 200×. The bigger fissures are visible

shows the structure of the central area of a sample compressed up to 3 180 MPa. The crushed out fragments and even the whole grains are visible. Their size is generally below twenty micrometers. The structure underwent evident loosening. The length of cracks between the grains was enclosed in the interval of 15÷50 $\mu$m, and many of them had a closed character (around single grain or group of grains). There occurred also bigger cracks of the character of fissures, as in the case of sample stressed up to 3 200 MPa – figure 11. The various areas of these samples demonstrated differentiated degree of structural degradation. In the central part of the sample compressed up to 3 180 MPa there were present even long cracks of the length of some hundreds of micrometers – Fig. 12. The most resistant of the investigated samples (compressing stopped at 3 804 MPa), showed a moderate degree of structure degradation. Even in its central part, the surface area of the crushed out grains did not exceeded 0.3 %. The mean length of cracks was about 20 $\mu$m and they were not longer than 50 $\mu$m. Structure of this sample was presented on the figures 7 and 13. The strength of the sample was the result of its homogeneous structure in the micro- and semi-macro scales.

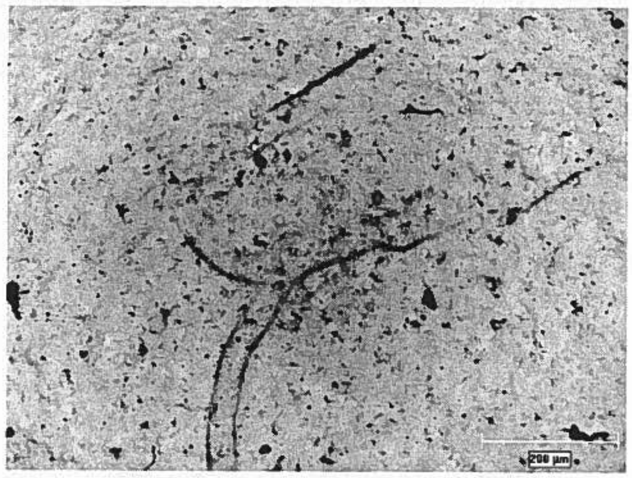


Fig. 12. Cracks in the central area of the sample loaded up to 3 180 MPa, magnification 100×

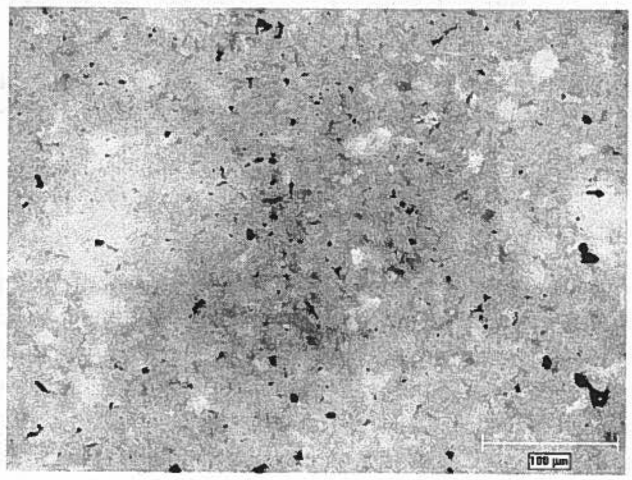


Fig. 13. Structure of the material in the border part of the most resistant sample. Its compressing was stopped at 3 804 MPa, magnification 200×

Observation of the structure of the samples compressed to advanced subcritical stage revealed presence of micro- and macrocracks – figure 14. Over 90 % of cracks underwent propagation along grain boundaries and only under 10 % within the grains. Points of initiation of the cracks were technological defects (gaseous and solid inclusions, fissures and partly broken grain boundaries), showed on figures 4 and 5. The earlier stages of AE signals were connected with propagation of these defects, which threshold energy was low. The most easily fracture bounds between bigger grains, especially elongated, and in perpendicular direction to compression. Centers of joined bigger grains are especially susceptible to destruction process. As a result the strength of samples containing centers and bands of the bigger grains ($10 \div 40$ $\mu$m) is considerably decreased. Cracking inside of bigger grains as well as at the boundaries of small grains is much less frequent (high threshold energy). During the critical stage of destruction, propagation of cracks occurs with very high velocity and throughout all elements of corundum structure. Damages of the structure, visible on figure 14, directly precede the critical stage of the material degradation.

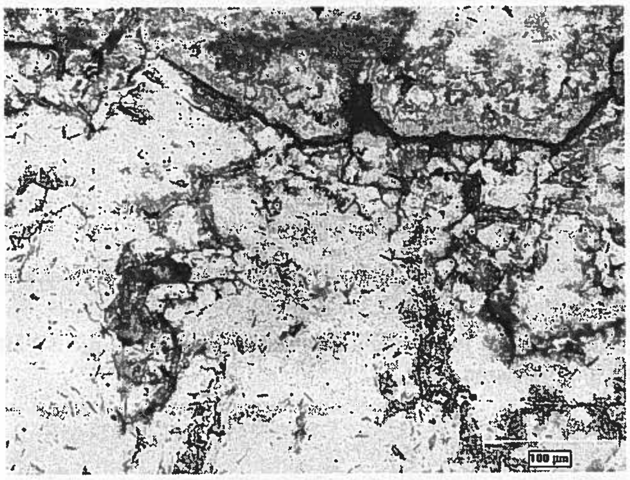


Fig. 14. Degradation of the structure visible on surface of the sample, which compression was stopped at 3 187 MPa, magnification 200×

## 4. Concluding remarks

Although there was used modern technology of making manufacturing of the samples, obtained structure of the material generally was not sufficiently homogeneous. Microscopic and ultrasonic investigations have revealed that the great considerable differences registered for the strength and the mechanical-acoustic characteristics of the samples are due to the faults of the material in the semi-macro as well as in the micro scales. Structure of the samples contained fine defects such as gaseous and solid inclusions, fissures and partly broken grain boundaries. These faults could have been introduced during technological process. Some defects, such as inclusions may perhaps be result of the properties of used granulated product. The distribution of the grains sizes demonstrated most often a bimodal form. Textural defects were connected with grains not uniformly distributed in the space. Sometimes they grouped into centres or bands. The occurrence of groupings of grains in the structure of the corundum material represents most probably the intermediate state, leading to the known effect of the abnormal grain growth (AGG) [10]. This phenomenon occurs most frequently in the oxide materials. It has a probabilistic character and its origin, despite many years of investigations, has not been sufficiently explained [11,12]. AGG effect occurs after a longer time of firing than in the case of the applied technology of the preparation of the samples. The quick increase of temperature in the course of thermal treatment favours its occurrence and such temperature raise – of the order of 200 °C by one hour – was realized. In the case of obtaining bigger and *longer time sintered* elements, the AGG effect would cause considerably greater differences in the size of the grains. The bigger grains grouped in centres or bands would join, attaining the size exceeding even 100 $\mu$m – figure 15.

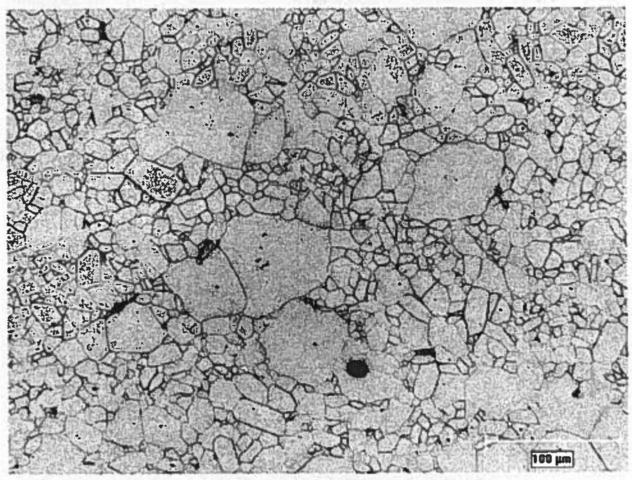


Fig. 15. Example of the abnormal grain growth effect in corundum material C 799 kind, magnification 200×

**Acknowledgements**

The paper was financially supported as part of the Research Project Nr N507 056 31/1289.

REFERENCES

[1] Z. Pohl – editor, High voltage overhead insulation in electrical power engineering (in Polish), Published by Wroclaw University of Technology (2003).
[2] IEC Publication 672-1:1995 Ceramic and glass--insulating materials, Part 1: Definitions and classification.
[3] IEC Publication 672-2:1999 Ceramic and glass--insulating materials, Part 2: Methods of tests.
[4] IEC Publication 672-3:1997 Ceramic and glass--insulating materials, Part 3: Specifications for individual materials.
[5] P. Ranachowski, F. Rejmund, A. Pawełek, A. Piątkowski, Acoustic emission in aluminous porcelain under compression load (in Polish), *Mat. XLIX*

*Open Seminar on Acoustics OSA 2002*, Warszawa-Stare Jabłonki, 465-472 (2002).

[6] P. Ranachowski, F. Rejmund, A. Pawełek, A. Piątkowski, Mechanical-Acoustic and Structural Investigations of Degradation Processes of Aluminous Insulator Porcelain C 130 Type, *Archives of Metallurgy and Materials* **52**, 4, 551-564 (2007).

[7] P. Ranachowski, F. Rejmund, A. Pawełek, A. Piątkowski, Structural and Acoustic Investigation of the Quality and Degradation Processes of Electrotechnical Insulator Porcelain under Compressive Stress, *AMAS Workshop on Nondestructive Testing of Materials NTM'03*, Warsaw, May 19-21, 179-196 (2003).

[8] A. G. Evans, T. G. Langdom, Structural Ceramics in: Progress in Mat. Sci. **21**, 171-441, Chalmers B., Christian J.N., Massalski T.M. (Editors), Pergamon Press, (1976).

[9] P. Ranachowski, F. Rejmund, A. Pawełek, A. Piątkowski, Investigation of Influence of Defectiveness in Aluminous Porcelain Structure on Fracture Process under Compressive Loading using Acoustic Emission Methods, *Archives of Acoustics* **31**, 4, 83-90 (2006).

[10] G.B. Prabhu, D.L. Bourell, Abnormal Grain Growth in Alumina-Zirconia Nanocomposites, *Nanostructured Materials* **5**, 6, 727-732 (1995).

[11] O.M. Ivasishin, S.V. Shevchenko, S.L. Semiatin, Modeling of Abnormal Grain Growth in Textured Materials, *Scripta Materialia* **50**, 1241-1245 (2004).

[12] P.R. Rios, Abnormal grain growth development from uniform grain size distributions due to a mobility advantage, *Scripta Materialia* **38**, 9, 1359-1364 (1998).

*Received: 12 March 2008.*