Investigation of the thermal expansion of the refractory materials at high temperatures

A Kostanovskiy, M Kostanovskaya, M Zeodinov and A Pronkin
Joint Institute for High Temperatures of the Russian Academy of Sciences (JIHT RAS), Izhorskaya 13 Bldg 2, Moscow 125412, Russia
E-mail: kostanovskiy@gmail.com

Abstract. We present the experimental investigation of the relative elongation and the coefficient of linear thermal expansion for monocrystaline alumina $\text{Al}_2\text{O}_3$ (1200 K – 1860 K), zirconia $\text{ZrO}_2$ (1200 K – 2730 K) and siliconized silicon carbide $\text{SiC}+\text{Si}$ (1150 K – 2500 K) in the specified range of temperatures. The following approach is used to measure the relative elongation: the through-cylindrical-marks located in the centre of isothermal part of the sample, and the measurement of temperature by two blackbody models, taken out of the area of the sample where the relative elongation is measured.

1. Introduction

Research interest for the new methods to measure relative elongation and true temperature of a sample appeared in the process of developing the State Primary Standard of the unit of the coefficient of linear thermal expansion (LTEC) for solids in the temperature range between 1500 K and 3000 K. It is known that at high temperatures the presence of temperature gradients at the ends of the experimental sample [1] may cause a methodological error when determining the relative elongation via the displacement of the sample ends. Therefore, in this paper we propose to measure the relative elongation not on the whole sample but on the segment where the isothermal approximation holds. Measuring temperature by means of absolutely blackbody (BB) models, which are applied to the same part of the sample as the measurement of relative elongation, might influence the measurement accuracy of the relative elongation. Therefore for measuring the sample’s true temperature we propose to locate BB models outside the sample segment at which the relative elongation is determined. Note, that measurements relative elongation and true temperature of a sample made of a semitransparent material are a non-trivial task. We choose a single-crystal aluminium oxide $\text{Al}_2\text{O}_3$, which is a semitransparent material meeting the requirements of standard measures of LTEC. $\text{Al}_2\text{O}_3$ has been in use by the leading metrological institutes all over the world for more than 40 years: e.g. at the Mendeleev All-Russia Research Institute of Metrology (VNIIM, Russia) in the temperature range of 90–1800 K [2, 3], as well as at the NIST (USA) in the temperature range of 90–2000 K [4]. Earlier to measure the relative elongation the following approach is used: the temperature of the $\text{Al}_2\text{O}_3$ sample is determined using a thermocouple, a resistance thermometer (given there is no contact with the sample), additionally an optical brightness pyrometer is used to measure temperature on the sample’s surface. Additional purpose of this research is to carry out an experimental determination of the relative elongation of a sample of $\text{Al}_2\text{O}_3$ measuring the “local” value of the true temperature using the...
BB models, situated inside the sample. Moreover, we investigate the thermal stability of the relative elongation as a function of the number of heating cycles. We report the results of the LTEC for zirconia ZrO$_2$ and siliconized silicon carbide SiC+Si too.

2. Experiment

Figure 1 schematically illustrates a facility, the main components of which are the specimen 1, the heater 2, the screens 3, the gas vacuum chamber 4, and the diagnostic system 5–9. The heater consists of a hollow cylinder made of graphite grade MPG-7, with diameters $d/D = 12.0/16.0$ mm, total length of 150 mm, and with an active length of 90 mm. The ratio of the active length of the heater to its inner diameter (90 mm/12.0 mm $\approx 10$) makes it possible to receive an isothermal field with a length of $\approx 15$ mm on an experimental sample. DC power supply maintains the preset rate of heating and cooling of the considered specimen, as well as the required holding time at maximum temperature. The power consumption of the heater is no higher than 18 kW. The diagnostic system to measure relative elongation consists of Sony NEX 5N digital camera 8, optical tube 7, and a Granit 11N lens 6. The camera is equipped with a 23.5 $\times$ 15.6 mm CMOS APSC sensor, 16.7 mega pixels. Sensitivity scale (at equivalent of ISO 100 with the use of an objective F 2.8 lens) is EV: 0–20. The specimen is placed horizontally on a dedicated holder made of graphite (grade GIP-4). The holder is fixed in the region of the isothermal segment of the heater at a single point - on a leg with a threaded joint, which minimizes a possible heating of the specimen by electric current. The specimen temperature is measured through a window of the chamber using an automatic micro pyrometer, with a sighting spot diameter of 0.4 mm and an operating wavelength of $0.65 \times 10^{-6}$ m. To determine the true temperature, we apply corrections for absorption in the glass of the chamber window and prism, and account for the perfection of geometrical dimensions of the BB model.

![Figure 1. Schematic diagram of the apparatus:](image)

Preliminary experiment enables to determine the length of the isothermal segment on the experimental sample. Preliminary experiment is conducted with a sample of the same material and size as the main experiment. This experiment reveals that the temperature drop is not higher than 2–3 K in the temperature range of 1200–2400 K at the length of 10 mm (which is comparable with the sensitivity of the pyrometer applied).

In preparation for the main experiment each specimen is exposed to the following procedures: the diameter in the central part and the ends is measured with a micrometer, the distance between the inner and outer points of the two marks is determined using a microscope installed at the mechanical stage.
with a fine adjusting screw, weighing is performed, then the specimen is annealed at 2500 K for approx \( \approx 30 \) min.

The specimen is placed into the chamber, which is filled with high purity He or Ar (see Table 1); the specimen position is adjusted, photographed in the cold state, and the distance between the centres of marks \( l_0 \) is determined (two marks it is photographed simultaneously). Then at a preset temperature variation rate \( dT/dt = 40 \) K/min, the specimen is heated up to temperature \( T_{\text{max}} \). The true temperature is measured in the BB models, which makes it possible to monitor fulfilment of condition \( T = \text{const} \) at the segment where the relative elongation is determined. Then the sample is photographed several times \( (n \geq 5 \text{ times}) \) with an interval of 5 min. Using preliminary processing of images, the time \( t_1 \) is determined within which the distance between marks is stabilized. The images acquired after \( t_1, n = 5 \) are further used to determine the distance between the marks \( l_{\text{max}} \). Then the specimen is cooled with the preset rate of temperature variation and held at \( T = 300 \) K till \( t_2 \). The heating-cooling cycles between \( T_{\text{max}} \) and \( T = 300 \) K are repeated. After the completion of a series of ten cycles, we again determine the distance between the marks and measure the diameter and mass of the sample at temperature \( T = 300 \) K. Determination of the geometrical dimensions and weight before and after the experiment makes it possible to monitor the absence of material loss.

The distance between the marks at ambient temperature before the experiment \( l_0 \) and relative elongation \( \Delta l/l_0 \) of the specimen in the heated state are determined by image processing. The lengths \( \Delta l = l_{\text{max}} - l_0 \) and \( l_0 \) are calculated in pixels. The images are prepared in Photoshop and processed in KOMPAS 3D. The distances \( \Delta l \) and \( l_0 \) are calculated by the centres of the marks, which are determined by three points inscribed in the mark circle. It is important that all images are acquired at a permanent focal distance and the centre of each mark is determined by the same points of the circle in both the cold and hot states. In order to identify the centre of each circle accurately, the images are processed at least five times.

3. Results

Results of research \( \text{Al}_2\text{O}_3 \). The experimental samples of \( \text{Al}_2\text{O}_3 \) are rods. The sizes of specimen and distance between marks are provided in Table 1. An anisotropic \( \text{Al}_2\text{O}_3 \) crystal was grown at the Ioffe Physicotechnical Institute using Stepanov’s method. The crystal belongs to the trigonal system, with spatial group \( 3 \text{m} \). The impurity content in the grown single crystals is of the order of \( 10^{-5} \) wt. %. The angle between the axis of the samples and the crystallographic C axis of the \( \text{Al}_2\text{O}_3 \) crystal is \( 90^\circ \). Four holes with a diameter of 1.8 mm are drilled into the sample perpendicular to the axis at distances of approximately 5 mm and 9 mm from the centre. The inner pair of openings is used as markers for measuring the elongation, while the outer pair is used to determine the temperature. Dampers (DE-24 grade graphite) are inserted into the outer openings. Holes of 0.7 mm in diameter and 4 mm deep are drilled in the dampers, the holes represent the BB models. Experiments with samples 1 and 2 are carried out in a gaseous medium of Ar at a pressure of 0.1 – 0.15 MPa. Experiments with sample 3 are conducted in a gaseous medium of He at the same pressure. Studying the effect of number of cycles on the relative elongation of sample 1 at \( T_{\text{max}} = 1246 \) K, 1442 K, 1640 K, 1859 K, we find that the mean value of relative elongation \( \Delta l/l_0 \) is nearly a constant when there is a change in the number of cycles \( N = 10 \). Heating up to a temperature \( T_{\text{max}} \) within the limits of the indicated relative expanded total uncertainty \( (U_{0.95} (\Delta l/l_0) \leq 21\% \) given a uniform distribution law and the coverage factor \( k = 1.65 \). Thus the mean value of relative elongation \( \Delta l/l_0 \) indicates stability of relative elongation \( \text{Al}_2\text{O}_3 \) for multiple heating cycles. A comparison of the two experiments, carried out on samples 1 and 2, shows that \( \Delta l/l_0 \) (for the same heating-cooling rates \( \approx 40 \) K/min, \( N = 10 \)) are reproduced. Note, that in the experiments sample 1 was treated to multiple thermal heating-cooling cycles at lower temperatures, while sample 2 was heated up to 1846 K without intermediate heat cycles at lower temperatures. In order to clarify the effect of the rate of change of temperature on the relative elongation, we carry out an experiment with sample 3, which is heated at a rate of \( dT/dt = 1.5 – 3.0 \) K/min up to the value of \( T_{\text{max}} = 1822 \) K and then cooled at the same rate. Using standard statistical test to compare two datasets with known variances under assumption of normal distribution we conclude that we can not reject the
null hypothesis of the equal mean of relative elongations for samples 1 \( (\bar{\Delta}l/\bar{l}_0)_1 \) and sample 2 \( (\bar{\Delta}l/\bar{l}_0)_2 \), thus we conclude that \( (\bar{\Delta}l/\bar{l}_0)_1 = (\bar{\Delta}l/\bar{l}_0)_2 \) are the same. Similarly, we find that \( (\bar{\Delta}l/\bar{l}_0)_1 = (\bar{\Delta}l/\bar{l}_0)_2 \approx (\Delta/l_0)_3 \) for sample 3. Hence, the rate of change of the temperature \( dT/d\tau = 1.5 – 4.0 \) K/min has no influence on the relative elongation for an investigated material. The dependence of LTEC \( \bar{\alpha} \) on temperature (a range 1220 K – 2730 K) for Al\(_2\)O\(_3\) in our experiments can be written as:

\[
\bar{\alpha} = \Delta l/(l_0(T_{\text{max}} - 293)) = 1.3377\times10^{-9}T + 6.377\times10^{-6} \text{[1/K].}
\]

The LTEC are identical to the data in [4] (where sample was oriented in a direction of 59\(^\circ\) to the trigonal axis of the crystal lattice) both in value and in the functional dependence from temperature.

### Table 1. Basic Geometrical Dimensions of the Experimental Samples and the Parameters of the Main Experiment.

| Sample number | Outer diameter, mm | Sample length, mm | Distance between the centers of the marks, mm | Rate of change of temperature, K/min | Gaseous medium |
|---------------|--------------------|-------------------|---------------------------------------------|-------------------------------------|----------------|
| 1. Al\(_2\)O\(_3\) | 5.65               | 30.2              | 9.93                                        | 40                                  | Ar             |
| 2. Al\(_2\)O\(_3\) | 5.66               | 30.5              | 9.99                                        | 40                                  | Ar             |
| 3. Al\(_2\)O\(_3\) | 6.4                | 30.2              | 10.03                                       | 1.5–3.0                             | He             |
| 4. ZrO\(_2\) | \(d/D=0.7/6.0\)    | 23.8              | 11.74                                       | 25                                  | He             |
| 5. SiC+Si     | 6.26               | 30                | 8.02                                        | 40                                  | He             |
| 6. SiC+Si     | 6.24               | 30                | 8.98                                        | 40                                  | He             |
| 7. SiC+Si     | \(d/D=2.0/6.4\)    | 28.3              | 8.94                                        | 40                                  | He             |

Results of research ZrO\(_2\). In this section we report results of experiments with ZrO\(_2\) (sample 4, Table 1). Phase structure of the material of the sample was defined via X-ray diffraction analysis using installation DRON-2. The analysis has shown, that the material consists of two modifications of zirconium oxide - tetragonal (80 volumetric percentages) and monoclinic (20 vol. %). The density of the sample of 5953.9 kg/m\(^3\) is slightly below the density of tetragonal modification ZrO\(_2\) - 6100 kg/m\(^3\) and above the density of monoclinic modification - 5700 kg/m\(^3\) [5]. The experimental sample is hollow cylinder with diameters of 0.7/6.0 mm and length 23.8 mm. Two holes with a diameter of 1.1 mm and a length of 2.65 mm are drilled at the distance of 11.74 mm between their centres in the middle part of the sample. The given apertures are used as the BB models to determine temperature during experiment and as marks when processing the photos to find the relative elongation. The material we study was created under conditions of formation of anion vacancies in the oxygen sublattice of ZrO\(_2\), therefore it possesses the expressed black color in the visible wavelength. During the experiment, pyrometer also works in a visible wavelength. Special check has shown, that the He-Ne beam of laser with wave length of 0.65 microns does not pass through the 0.8 mm thick disk. Hence the studied material is opaque at the specified length of a wave. Secondly, the investigated material is characterized by low values of heat conductivity [5]. The combination of low heat conductivity and thickness of a sample’s wall of 2.65 mm inevitably should lead to rather high temperature drops along the radius. This effect generates the need to account for the degree of perfection of the BB model. We apply the technique by Sparrow [6] for BB models in which the temperature variation along the circular cylindrical cavity is described by the linear relationship. The
mean of the temperature at the bottom of the cavity and the temperature at the cavity opening is used as the defining temperature of the relative elongation $\Delta l_0$ and LTEC coefficient. To find an average value of relative elongation, we repeat the experiment three times ($N = 3$) for each value of $T_{\text{max}}$. The dependence of LTEC $\bar{\alpha}$ on temperature for the investigated material ZrO$_2$ (in the range between 1200 K – 2730 K) is then given as:

$$\bar{\alpha} = \Delta l/(l_0(T_{\text{max}} - 293)) = 1.0349 \times 10^{-9} T + 1.255 \times 10^{-5} \text{ [1/K]}.$$  

(2)

The resulting LTEC $\bar{\alpha}$ is an increasing function of temperature and coincides with values known from literature for temperatures between 1200 K - 1800 K [5, 7]. The experiments in this paper expand the range of the studied values of $T$ for almost 1000 K above the temperatures. Results of research SiC+Si. Samples SiC+Si represent cores of round section (samples 5, 6) and the hollow thin-walled cylinder (sample 7 see Table 1). The three samples have been produced by company “Podolskogonepor” using the method of cold plastic formation and the subsequent high-temperature (2400 K) sintering. The mass fraction of SiC+Si is above 97%, the porosity of no less than 30%. In each sample we make four holes by the method of electroerosive processing using NPK “DELTA-TEST”. The two internal holes (marks) are created perpendicularly to the axe of the sample and go through the sample of diameter 1.2 mm. Two additional holes (representing the BB model) are made in the direction away from the centre of the marks at the distance of $\approx 2$ mm; the holes are of diameter of 1.1 mm and depth of $\approx 4$ mm (samples 5, 6); for sample 7 the depth of the hole equals to the thickness of the sample’s wall. Experiments are conducted with the He environment. Experiment and technique of processing correspond to experiments with ZrO$_2$. Thus, the LTEC of the SiC+Si between 1150 K and 2500 K is given by the expression:

$$\bar{\alpha} = \Delta l/(l_0(T_{\text{max}} - 293)) = 2.5344 \times 10^{-9} T + 2.761 \times 10^{-6} \text{ [1/K]}.$$  

(3)

The overall relative expanded uncertainty $U(\Delta l/l_0)$ does not exceed 21% at $T_{\text{max}} = 1250$ K and 12% at $T_{\text{max}} = 1850$ K for all experiments (distance between of the centres of the marks $\sim 10$ mm). The contribution of $l_0$ to $U(\Delta l/l_0)$ is approximately 70%, while the contribution of $l_{T_{\text{max}}}$ is about 30%.

4. Conclusion

In the paper we present the experimental results of the coefficient of linear thermal expansion for Al$_2$O$_3$, ZrO$_2$, SiC+Si at high temperatures. We put before ourselves a problem to increase accuracy of definition of the LTEC for samples of small length. Following approach are used: the through-cylindrical marks located in the centre of isothermal part of the sample and the measurement of temperature by two BB models, taken out of the area of the sample where the relative elongation is measured.

Acknowledgements

This study is supported by the Russian Foundation for Basic Research, project no. 15-08- 06279.

References

[1] Petukhov V and Chekhovskoi V 1972 High Temp. - High Pressures 4 671-677
[2] Amatuni A N et al 1997 Tables of Standard Reference Data. Materials for Standard Measures of the Coefficient of Linear Thermal Expansion. Single-Crystal Aluminum Oxide. Coefficient of Linear Thermal Expansion GSSSD No. 176-96 (Moscow: Standartov)
[3] Kompan T A et al 1999 Tables of Standard Reference Data. Materials for Standard Measures of the Coefficient of Linear Thermal Expansion. Single-Crystal Aluminum Oxide with 59° Orientation Relative to the Trigonal (C) Axis of the Crystal Lattice GSSSD No. 186-99
(Moscow: Standartov)

[4] Hahn T A 1978 Thermal expansion of single crystal sapphire from 293 to 2000 K Standard reference material 732 AIP Conf. Proc. Thermal Expansion – 6 (N-Y, London) p 191

[5] Touloukian Y (Ed.) 1967 Thermophysical Properties of High Temperature Solid Materials (New York: Macmillan)

[6] Sparrow E 1965 Appl. Opt. 4 41-43

[7] Haggerty R, Sarin P, Apostolov Z, Driemeyer P, Kriven W 2014 J. Am. Ceram. Soc. 97 2213.