Physical and FE-Simulation of Thermal Shock Behaviour of Refractory Ceramics

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Thermal shock resistance of refractory ceramics results from the complex interaction between the stress and thermomechanical properties of the material. To characterize thermomechanical properties of refractory ceramics, physical simulation was carried out using the simulation and test centre Gleeble 3500. A new heating method based on the heat transfer from electrical heater into the specimens was developed to test non electrical conductive materials like alumina-mullite refractory ceramic. The graphite containing refractory ceramics were heated directly via electrical current. In addition, for enhanced understanding of damage and failure mechanisms under thermal shock loading, a combination of experimental testing and numerical simulation methods has been used. The thermal shock behaviour of the alumina (99.7 %) disk samples has been investigated by using a plasma test stand: the bottom of the ceramic disks were locally heated in the centre by plasma beam; during the heat treatment the temperature distribution at the top of the sample was recorded with a thermographic system. To characterise the thermal shock resistance, a thermomechanical simulation was subsequently carried out. It calculates the temperature and stress distribution within the ceramic disks. Beside reference material alumina (99.7 %), more refractory ceramics fabricated within other subprojects of SPP 1418, such as Ca-Aluminate/Alumina-Multilayer (Fraunhofer IKTS), Multilayer Laminates (WW3, FAU), Refractory Castables Containing Eutectic Aggregates (GHI, RWTH), have been investigated using this plasma test stand. By means of investigation and comparison of the damage mechanisms under thermal shock loading, refractory ceramics were approved, which exhibit better thermal shock resistance and are suitable for steel casting applications.

1 Introduction

The Priority Programme SPP 1418 Feuerfest – Initiative zur Reduzierung von Emissionen – FIRE is a research programme with the main objective of the development and characterization of carbonless refractory ceramics based on the oxides. It is funded by the German Research Foundation (DFG). The present work in project area testing technology within the framework of SPP 1418 focuses on the development of a combined testing method based on physical and thermomechanical numerical simulation to study the thermal shock behaviour of refractory ceramics.

A physical simulation and test centre, Gleeble 3500 (Dynamic Systems Inc./US), is widely used for the thermomechanical characterization of materials including metals and alloys [1]. As to the authors knowledge no exact study on the thermomechanical properties of refractory ceramics using Gleeble has been reported. Al$_2$O$_3$–C refractory ceramics are widely used in metallurgical industry due to their outstanding properties such as thermal shock resistance, corrosion and erosion resistance, spalling resistance and high strength [2, 3]. Carbon/graphite additives can be used to improve the thermal stability of the refractory ceramics owing to their high thermal conductivity, low thermal expansion and low wettability by liquid metals [4, 5]. In the present work, therefore, the thermomechanical properties of Al$_2$O$_3$–C refractory ceramics were investigated using Gleeble 3500.

Compression, dilatometer and thermal cycling tests were performed. The measured compressive strength and thermal expansion for different temperatures, heating, and strain rates are reported. Thermal fatigue behaviour can be determined by comparing the µCT results before and after thermal cycling tests. In addition, carbon-free mullite ceramic materials bear a real potential for CO$_2$ emission reduction compared to carbon containing refractories with high CO$_2$ emissions. Mullite refractory material containing...
an alumina core and a mullite shell was developed [6], which was expected to have a better high temperature performance as compared to plain mullite. Furthermore, the thermal behaviour of the alumina-mullite samples was studied [6]. Thermal cycle tests for the fabricated hollow cylinders were carried out using the test- and simulation centre Gleeble 3500 [6]. The main application of the refractory ceramics is in the steel casting, whose thermal shock resistance is applied only from one side. A suitable technique is the generation of a high temperature gradient via lamp or laser irradiation [7] or via plasma torch [8], which was applied in this study. In contrast to the established techniques of water and air quenching, this test gives a result, which allows a statement concerning the behaviour of a refractory in its real application. Therefore, thermal shock behaviour of refractory ceramics including the reference material alumina (99.7 %) [8] and refractory ceramics developed and fabricated within other subprojects of SPP 1418 were experimental investigated using this developed plasma test stand; the temperature distribution at the reverse side of the sample was measured in situ with a thermographic system [9]. Numerical thermo-mechanical simulations were subsequently carried out to describe the temperature and stress distributions in the tested samples.

2 Experimental procedure and FE-simulation

2.1 Alumina-graphite refractory ceramics

(Cooperation with Institute of Ceramics, Glass and Construction Materials, TU Bergakademie Freiberg)

A novel Al$_2$O$_3$-C refractory ceramic was fabricated by using commercial grades of alumina and graphite. The preparation of the alumina-graphite samples was described in detail elsewhere [2]. Two types of alumina-graphite samples were used in the present study which were labelled as A1-S10 and A1G-S10. The second composition A1G-S10 is a granulated type of the first composition A1-S10. Electrical conductive samples Al$_2$O$_3$-C refractory ceramic used in the present work can be heated directly by the Joule effect using the Gleeble 3500. Fig. 1a shows the compression sample fixed into the Gleeble 3500. The sample is a solid cylinder with a diameter of 10 mm and a length of 15 mm. Compression tests were performed at room temperature, 1000 °C and 1300 °C and different strain rates of 10$^{-3}$, 10$^{-2}$ and 10 s$^{-1}$, respectively under Ar atmosphere. In addition, before and after the thermal cycle testing the cylinders were analysed using the microcomputed tomography (μCT) (Micro CT 1172, SKYSCAN/BE). The thermal fatigue behaviour can be determined by comparing the μCT results before and after thermal cycling tests. Dilatometer tests were carried out from room temperature up to 1500 °C using different heating rates of 100 °C/s and 300 °C/s.

2.2 Alumina-mullite ceramics

(Cooperation with Institute of Materials and Joining Technology, Otto-von-Guericke University of Magdeburg)

The preparation of the alumina-mullite samples was described in detail elsewhere [6]. Hollow cylinders were obtained from the sintered samples following a machining process, which were used to investigate the thermal behaviour. These electrically non-conductive samples cannot be heated by Joule effect in the Gleeble 3500 machine. Therefore, a heater unit was developed in this study in order to heat the electrically non-conductive hollow cylinder samples, as shown in Fig. 1b. A maximum temperature of 800 °C in the hollow cylinder can be achieved. The hollow cylinders were analysed before and after thermal cycling using μCT [6].

2.3 Reference material: alumina (99.7 %) disks with different notches

A test stand was developed to investigate the thermal shock behaviour of ceramics, which was described in detail elsewhere [8]. A combination of experimental testing and FE-simulation methods was used to investigate the thermal shock behaviour of alumina (99.7 %) disk samples without notch [8]. The disks were rapidly heated in the centre using a plasma beam resulting in temperature and thermal stress gradients, which led to the failure of the samples. A finite element model has been developed and approved by means of comparison with experiments [8]. In addition, wide research focused on the investigation on the thermal shock behaviour of the alumina (99.7 %) disks with notches using the developed plasma test stand. Alumina specimens are 2 mm thick and have a diameter of 40 mm. Three types of notch length (3, 6, and 9 mm, respectively), and two types of notch (U-type and V-type) were prepared for thermal shock testing in the plasma test stand.

2.4 Refractory ceramics based on the calcium-aluminate/alumina-multilayer

(Cooperation with Fraunhofer IKTS)

The preparation of refractory ceramics based on the calcium-aluminate/alumina-multilayer was described in detail elsewhere [9]. Thermo shock behaviour of fabricated calcium-aluminate/alumina-multilayer with different porosity was investigated in the plasma test stand [9].

2.5 Multilayer laminates based on alumina and alumina/zirconia

(Cooperation with Chair of Glass and Ceramics, University of Erlangen-Nuremberg)

The fabrication of multilayer laminates was described in detail elsewhere [10] whose thermal shock resistance was investigated in the plasma test stand. Square samples with side length of 40 mm and thickness
in the range from 6 – 10 mm due to different fraction and particle size of applied powders were used for the thermal shock testing. The bottom of the ceramic samples was locally heated at the centre using the plasma torch. The heating lasted 3 min. In this study the electrical current of 60 A was used. After the thermal shock testing the multilayer laminates were analysed at the Chair Glass and Ceramics, University Erlangen-Nuremberg, in order to visualize the damage mechanisms including possibly formed cracks and delaminations.

2.6 Refractory castables containing eutectic aggregates
(Cooperation with Chair of Ceramics and Refractory Materials, RWTH Aachen)
The fabrication of refractory castables containing eutectic aggregates was described in detail elsewhere [11] whose thermal shock resistance was investigated in the plasma test stand. Disk samples with diameter of 50 mm and thickness of 9 mm were used for the thermal shock testing. The bottom of the ceramic samples was locally heated at the centre using the plasma torch. The heating lasted up to 3 min. In this study the electrical current of 60 A was used. After the thermal shock testing the refractory castable samples were analysed at the Chair Ceramics and Refractory Materials, RWTH Aachen, in order to evaluate the mechanisms of the crack initiation and propagation after a defined thermal shock cycle and in order to investigate the effect of aggregates on the crack path and to understand the energy consumption mechanisms by the thermal shock testing.

3 Results and discussion
3.1 Alumina-graphite refractory ceramics
(Cooperation with Institute of Ceramics, Glass and Construction Materials, TU Bergakademie Freiberg)
The dependence of compressive strength on the strain rate is shown in Fig. 2a for room temperature and 1000 °C. At room temperature, the compressive strength of the sample A1G-S10 increases slightly from 26.5 ± 1.2 MPa to 30 ± 1.5 MPa with increasing strain rate from 10^{-4} – 10^{-1} s^{-1}. At room temperature, it was found that the lower strain rate such as 10^{-4} s^{-1} and 10^{-2} s^{-1} has no considerable effect on the measured compressive strength; while the higher strain rate of 10^{-1} s^{-1} has more effect on the compressive strength. The compressive strength at room temperature of the sample A1-S10 increases with higher strain rate, which is higher than that of the sample A1G-S10 for all the tested strain rates. At the testing temperature of 1000 °C an increase in compressive strength of the sample A1-S10 increases with higher strain rate, which is higher than that of the sample A1G-S10 for all the tested strain rates. At the testing temperature of 1000 °C an increase in compressive strength of the sample A1-S10 was observed with increasing strain rate from 10^{-4} – 10^{-1} s^{-1}. At high temperature, the compressive strength of the sample A1G-S10 decreases firstly and then increases slightly when the strain rate increases from 10^{-4} – 10^{-1} s^{-1}. Lankford reported the strain rate dependence of compressive strength in ceramics Al_{2}O_{3}, SiC and...
At the lower strain rate ($10^{-4}$ to $10^2$ s$^{-1}$) the compressive strength of Al$_2$O$_3$ at room temperature increases slightly with increasing strain rate, while that of SiC remains nearly constant [12]. In this case the relation between compressive strength and strain rate can be described by the following equation

$$\sigma \propto \varepsilon^{(1+c_n)}$$

where $\sigma$ is compressive strength, $\varepsilon$ is strain rate and $c_n$ is a value ranging from 50 – 200 [12, 13].

The dependence of compressive strength on the testing temperature is shown in Fig. 2b. All illustrated samples were tested at a constant strain rate of $10^{-2}$ s$^{-1}$. The compressive strength increases with higher testing temperature. In this case the testing temperature has a considerable effect on compressive strength of the alumina-graphite samples. Under the strain rate of $10^{-2}$ s$^{-1}$ the measured compressive strength of the sample A1G-S10 is higher than that of the sample A1-S10. This is attributed to the higher open porosity of the granulated refractory composition, originated from the higher binder amount. The maximum of compressive strength at high temperature for refractory ceramics can be performed due to the tension in the structure based on the different thermal expansion of the material components [14].

The thermal expansion of alumina and graphite at 1000 °C is 0,8 % and 0,2 – 0,28 %, respectively [14]. Therefore, in this work the enormous difference of the thermal expansion between alumina and graphite resulted in a maximum compressive strength at 1000 °C. The compressive strength decreases slightly at 1300 °C (Fig. 2c). The cylinder surface after the compressive testing is shown in Fig. 2b, which was carried out under a strain rate of $10^{-4}$ s$^{-1}$ at the temperature of 1000 °C. It was found that the fracture mode is predominantly intergranular. The compressive stress-strain curves tested at different temperature with a strain rate of $10^{-4}$ s$^{-1}$ are shown in Fig. 2c. The compressive stress-strain curves derived from non-linearity, which is a typical stress-strain curve of structurally flexible refractory [14], due to the addition of graphite. After reaching the maximum stress, the samples tested at high temperature of 1000 °C and 1300 °C still remained its integrity and it did not fail, as shown in Fig. 2c 2) and Fig. 3). It indicates that the fabricated alumina-graphite refractory ceramic under compressive loading (strain rate $10^{-4}$ s$^{-1}$) at high temperature of 1000 °C and 1300 °C exhibited no catastrophic brittle fracture. It was found that a “quasiplastic” fracture mode occurred during the compressive testing under low strain rate of $10^{-2}$ s$^{-1}$ and the plastic deformation zone increases with increasing test temperature. It should be pointed out that, no decrease in compressive strength of the sample A1G-S10 was observed considering the standard deviation which was measured after 5 thermal cycles (Fig. 3). The comparison of the $\mu$CT-images before and after thermal cycles exhibit no considerably thermal crack or damage in the cylinder, as shown in Fig. 3a-b.

The cross expansion of the cylinder sample were determined during temperature cycles up to 1500 °C (Fig. 2d). The average linear coefficients of thermal expansion in the temperature range of 22 – 1500 °C, calculated from the slopes of these curves, is $6,9 \times 10^{-6}$ 1/°C; the thermal expansion is independent of the heating rate, as shown in Fig. 2d.

3.2 Alumina-mullite ceramics

(Cooperation with Institute of Materials and Joining Technology, Otto-von-Guericke University of Magdeburg) [6]

The results of heated hollow cylinders in the Gleeble 3500 and relevant discussion were described in detail elsewhere [6].

3.3 Reference material: alumina (99,7 %) disks with different notches

The V-type notches have an additional 1 mm very thin and sharp crack on the tip of the notch. These V-type notch models are only studied in sake of the theoretical analyses, as shown in Fig. 4b. Maximum temperature exists on the bottom side of the specimens where the specimens are heated. It is also detected that the value of maximum temperatures on fracture moment decreases with longer notch lengths, as shown in Fig. 4c. In light of this information it can be said that with longer notch lengths the specimen reach their fracture strength with lower values of temperatures. The calculated stress distributions on each specimen and an overlook at the specimen are displayed in Fig. 4d–f. Observing the stress distributions on each specimen, it can be seen that on each specimen there exists...
a compression section in the middle area of the disc shaped specimen [15]. Regarding this finding it can be assumed that with a very long crack/notch length if the tip of the crack is located in the compression area the crack propagation would not be catastrophically. As seen in the practical experiments of 9 mm long notch lengths, even though after the thermal shock effect, there occurs crack propagation that the specimen does not dismember itself. Another important section of the developed stress distributions is that in near crack tip area the tangential stress values increase to approximately 350 MPa. This increment of tangential stresses causes the start of the crack propagation on the specimen.
3.4 Refractory ceramics based on the calcium-aluminate/alumina-multilayer

(Cooperation with Fraunhofer IKTS) [9]
The results of investigation on the refractory ceramics based on the calcium-aluminate/alumina-multilayer and relevant discussion were described in detail elsewhere [9].

3.5 Multilayer laminates based on alumina and alumina/zirconia

(Cooperation with Chair of Glass and Ceramics, University of Erlangen-Nuremberg)
Total ten different multilayer laminate types were tested in the plasma test stand. By way of example, the measured temperature distributions on the top of a multilayer laminate type are shown in Fig. 5 for different time steps of the thermal shock testing. Recorded thermographic image during thermal shock testing is shown in Fig. 5a. A side view of this tested multilayer laminate is shown in Fig. 5b. A crack could be detected which propagated through the entire cross section. Pronounced damage including macro-cracks could be found on the top of this laminate type after thermal shock testing, which only consists of bimodal layers. The higher Young’s modulus and high thermal conductivity through all bimodal layers resulted in crack propagation throughout the entire cross section. More results and discussions will be published in a corporate publication with Chair of Glass and Ceramics, University of Erlangen-Nuremberg, and Institute of Energy and Climate Research – 2, Forschungszentrum Jülich GmbH in the foreseeable future.

3.6 Refractory castables containing eutectic aggregates

(Cooperation with Chair of Ceramics and Refractory Materials, RWTH Aachen)
Total one reference refractory castable type and six refractory castable types containing different aggregates were tested in the plasma test stand. By way of example, the measured temperature distributions on the top of a refractory castable are shown in Fig. 6 for different time steps of the thermal shock testing. Recorded thermographic image during thermal shock testing is shown in Fig. 6a. A crack could be detected which propagated from edge to the centre during the defined thermal shock cycle (Fig. 6), as shown in Fig. 6a. Crack path and damage mechanisms were investigated in detail using scanning electron microscope (SEM) at the Chair of Ceramics and Refractory Materials, RWTH Aachen. By means of investigation and comparison of the crack propagation path and damage mechanisms of different refractory castables it is tried to detect the effect of the applied aggregates on the fracture energy consumption subject to thermal shock treatment. More results and discussions will be published in a corporate publication with Chair of Ceramics and Refractory Materials, RWTH Aachen, in the foreseeable future.

4 Conclusions
Thermomechanical properties of alumina-graphite refractory ceramics were investigated using physical simulation and test centre Gleeble 3500. The dependence of compressive strength on the strain rate and temperature was studied. At room temperature, compressive strength increases slightly with increasing strain rate in the range of $10^{-4}$ – $10^{-1}$ s$^{-1}$. An increase in compressive strength was observed with increasing test-
ing temperature. The stress-strain curves under compressive loading (strain rate $10^{-4}$ s$^{-1}$) at high temperature of 1000 °C and 1300 °C derived from non-linearity, which is a typical stress-strain curve of structurally flexible refractory. The average linear coefficients of thermal expansion in the temperature range of 22 – 1500 °C, calculated from the slopes of these curves, is $-6.9 \times 10^{-6}$ 1/°C; the thermal expansion is independent of the heating rate.

The thermal behaviour of novel alumina-mullite refractory material was investigated using the physical simulation and test centre Gleeble 3500. A heater unit was developed in order to heat the electrical non-conductive hollow cylinder samples. The thermal fatigue behaviour was determined by comparing the µCT results before and after thermal cycle tests. Fatigue crack initiation and propagation due to the thermal cycling were detected by µCT-analysis [6]. A test stand was developed in order to study the thermal shock behaviour of the refractory ceramics [8]. Beside reference material alumina (99.7 %), more refractory ceramics fabricated within other subprojects of SPP 1418, such as Ca-Aluminate/Alumina-Multilayer (Fraunhofer IKTS), Multilayer Laminates (WW3, FAU), Refractory Castables Containing Eutectic Aggregates (GHI, RWTH), have been investigated by using this plasma test stand, in order to understand the damage and failure mechanism under thermal shock loading.

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**References**


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