MECHANICAL BEHAVIOR OF CARBON-BONDED MAGNESIA (MgO-C) AT TEMPERATURES UP TO 1500 °C

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ABSTRACT

Carbon-bonded magnesia (MgO-C) with maximal magnesia grain sizes of 2 mm and a graphite content of 10 % was investigated regarding its mechanical properties. Therefore bending tests and fracture mechanical tests were carried out at temperatures up to 1500°C under inert atmosphere. The results showed change of mechanical behavior in dependency of temperature. Thus, the results indicated that bending strength of MgO-C increases with increasing test temperature up to 1300°C. An increase in fracture toughness of the material with increasing test temperature was also observed in fracture mechanical tests on single edge notched bend samples (SENB). The tests were complemented by analysis of microstructure, density and porosity.

INTRODUCTION

In all industrial areas where high-temperature processes occur, lining of the equipment with refractories is required. Thus, refractory materials represent an indispensable material group for a large part of the industry such as metallurgy and energy production. The refractory lining of metallurgy vessels in steel production is subjected to very high temperatures up to 1750°C as well as extreme chemical and mechanical stresses by metal melts and slag, respectively. In converters, electric furnaces and steel ladles carbon-bonded refractories are particularly suitable, since they have very good thermal shock resistance[1]. To improve carbon-bonded refractory materials and for development of new material solutions, general understanding of the mechanical behaviour is necessary. Therefore, the aim of the present work is to investigate the bending strength and fracture toughness of carbon- bonded magnesia at room temperature as well as high temperatures. For testing a machine with inductive heating device and an inert gas chamber was used with four-point bending setup.

EXPERIMENTAL DETAILS

Processing of samples

For material preparation electrofused MgO (grain diameter up to 2 mm), phenolic resin and graphite flakes (purity: 94 %) were mixed in an Eirich Mixer. Afterwards, the mixture was pressed uniaxially at 100 MPa into bending bars (25 mm x 25 mm x 145 mm). After pressing, the bars were heated to 180°C for polymerisation of the bonding agent and coked under reducing atmosphere at 1000°C in a coke bed afterwards. More details are given by Solarek et al.[2].

For the fracture mechanics tests the samples were notched prior to coking (see Fig. 1). For this, a single pre-notch was first introduced into the samples with a width of 0.5 mm (c) and a depth of approx. 10 mm (b) with a hard metal saw. Then the notch was honed using a razor blade to get a sharp notch with a length of ≥ 0.5 mm und an average diameter of the notch root of < 20 µm. An example for the finished notch in SENB specimen is shown in Fig. 1.

For the determination of the Poisson´s ratio small cylinders (Ø 25 mm x 25 mm) were turned out of a bar prior to coking.

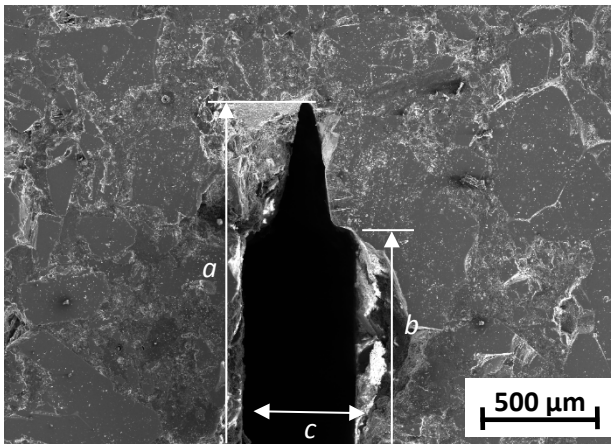


Fig. 1: Finished notch in SENB specimen, scanning electron microscopy.

Methods

The microstructure was characterised with a field emission scanning electron microscope (MIRA 3 XMU, Tescan). Therefore, specimens were grounded and polished up to a particle size of 1 µm and coated with a thin layer of carbon subsequently. For investigation of porosity and density, the Archimedes principle was applied with toluene (C_7H_8) as a media to prevent hydration of magnesia. The true density was determined with a pycnometer (AccuPyc 1350).

The Poisson´s ratio was determined by a compression test on one cylindrical sample at room temperature. For the measurement of sample elongation in the longitudinal and transverse direction, the specimen was covered with four gauges around the circumference transversely and longitudinally to the load direction. Then the sample was loaded with almost 20 MPa with a rate of 200 N/s. The Poisson´s ratio was calculated using equation (1).

$$\mu = \frac{\varepsilon_{\text{transversal}}}{\varepsilon_{\text{longitudinal}}} \quad (1)$$

For bending tests and fracture mechanic tests, the loading axis was parallel to pressing direction. Bending tests and fracture mechanics tests were carried out in four-point bending using a 20 kN electro-mechanical testing machine (Z020, Zwick) equipped with an inert gas chamber (Maytec) and an inductive heating unit (TrueHeat HF 5010, Hüttinger) with copper coils. For testing, the chamber was evacuated and filled with argon twice. During the tests, the temperature was measured with a pyrometer (MS 09, Sensortherm) using an emission coefficient of 0.92. In contrast to earlier high-temperature tests on MgO-C[2] a layer of graphite spray was applied on each sample before start the experiments. Thus, the temperature measurement was not affected by the different emission coefficients of the MgO- and graphite-grains and temperature measurement took place under same conditions for each test.

The bending tests were carried out at room temperature, 700°C, 850°C, 1000°C, 1150°C, 1300°C, and 1450°C on non-notched samples. Two samples were tested at each test temperature. The samples were loaded with a rate of 50 N/s until failure. Strain measurement was performed with a high-temperature

strain gauge with alumina rods. By means of the maximum load of the load-deflection-curve, the bending strength σ_b was calculated using equation (2), where F is maximal load, L is the outer span and l the inner span of the four-point bending setup, h is sample height and w is sample width.

$$\sigma_b = \frac{3F(L-l)}{2wh^2} \quad (2)$$

The fracture mechanical tests were performed at room temperature and 700°C – 1300°C in 100 K intervals. One sample was tested at each test temperature. Therefore, the samples were loaded with a rate of 0.1 mm/min until failure. During the tests, the applied load was measured with a load cell. In order to measure the displacement of the notch under load, Al_2O_3-distance pieces were placed on the bottom of the SENB-specimens as shown in Fig. 2. The crack opening was determined from the increase of the distance between the Al_2O_3-pieces which was measured using an optical laser-measurement system (LS-7070M, Keyence).

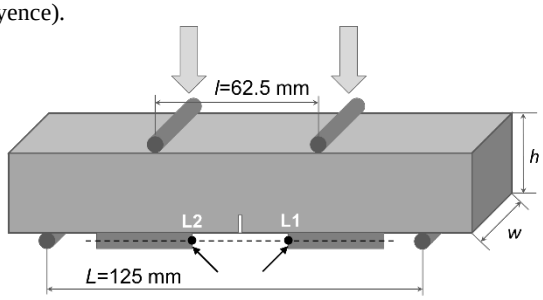


Fig. 2: Experimental set up for four point bending test of SENB, with distance pieces and laser line (dashed black line) of the optical measurement system.

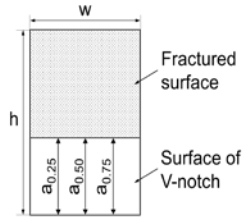
Transmitter and receiver of the optical measurement system were installed in front of the viewing window of the inert gas chamber. The transmitter of the optical measurement system was aligned, so that the laser line hit the distance pieces, as shown in Fig. 2 (dashed line). A mirror placed behind the sample threw the laser light back to the receiver. The inner edges of the distance pieces served as reference for the measurement, as shown in Fig. 2 (black arrows). As a result, the distance between the two point L1 and L2 increased as the load increased, whereby the crack opening can be measured indirectly.

After failure, microscopic images of the fracture surfaces were taken. The fracture toughness K_{Ic} was calculated using equation (3), where α is the relative depth of the notch, Y^* a geometrical factor and F is the maximum load that can be taken from the load-displacement curve[3].

$$K_{Ic} = \frac{F}{w\sqrt{h}} \cdot \frac{L-l}{h} \cdot \frac{3\sqrt{\alpha}}{2(1-\alpha)^{3/2}} \cdot Y^* \quad (3)$$

$$Y^* = 1.9887 - 1.326\alpha - \frac{(3.94 - 0.68\alpha + 1.35\alpha^2) \cdot \alpha(1-\alpha)}{(1-\alpha)^2}$$

The relative notch depth α was determined using equation 4 where $\bar{a}$ is the mean of the depth of the V-notch. For determination of $\bar{a}$ the fractured surface was measured by an optical microscope as shown in Fig. 3.



$$\alpha = \frac{\bar{a}}{h} \quad (4)$$

$$\bar{a} = \frac{a_{0.25} + a_{0.5} + a_{0.75}}{3} \quad (5)$$

Fig. 3: Schematic illustration of a fractured specimen showing the points for determination of the length of the notch[3].

RESULTS

Microstructure

Fig. 4 shows the microstructure of the material in BSE-contrast. It consists of different sized magnesia grains, embedded in a carbon-rich matrix. Within the coarse magnesia grains, CaO/SiO_2 impurities were observed, mostly on the grain boundaries (see dashed arrows). The microstructure reveals cracks within the large magnesia grain (see solid arrows), which were probably formed during shaping of the samples at 100 MPa. In addition, cracks within the matrix and between matrix and magnesia grains were observed. These cracks are the result of different thermal expansion coefficients of magnesia and graphite and are generated during cooling from the coking temperature[4, 5]. After coking, specimens exhibited an open porosity of approx. 13 %, a closed porosity of approx. 2 %, a bulk density of 2.9 g/cm^3 and a theoretical density of 3.4 g/cm^3.

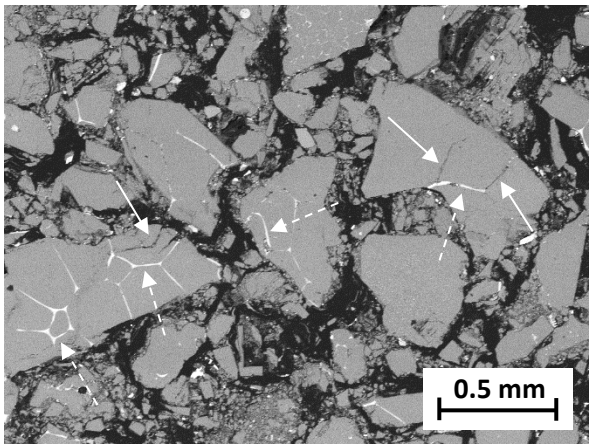


Fig. 4: Microstructure of carbon-bonded magnesia, scanning electron microscopy in backscattered electron contrast.

Mechanical tests

Poisson´s ratio and bending tests

The Poisson's ratio $\boldsymbol{\mu}$ determined in compression was 0.48. This matches with the Poisson´s ratio of Hino and Kiota[6] of 0.42 determined on MgO-C coked at 1350°C.

Based on the load-displacement curves from the bending tests, the bending strength of MgO-C was determined at different test temperatures using Eq. 2. Fig. 5 shows the development of bending strength with increasing test temperature. Thus, the average bending strength at room temperature is 2.53 MPa and rises to 4.81 MPa at 1300°C. Above 1300°C the bending strength decreases. Franklin et al.[4] reported an increase of Young´s modulus and strength with temperature as well for MgO-C. Mason and Knibbs[7] observed an increase in the Young´s modulus of graphite with increasing temperature. They explained this phenomenon with closure of cracks within the matrix due to different coefficients of thermal expansion and connected the maximal strength to the cooking temperature. Consequently, the stiffness and strength of the material increase. In general, the strength of graphite increases with increasing temperature, the maximum of the strength is expected at 2500°C[8]. Baudson[5] observed an increase in the Young´s modulus of MgO-C with growing test temperature, too. They also attributed this to closure of cracks and pores in the material as well as to a compressing of it, because of sintering of the MgO grains at high temperatures. Despite the increasing strength of MgO-C with increasing test temperature, the material strength decreasing above 1300°C. This can be explained by softening due to thermally activated deformation (creep). Former tests on the same material have shown a strong increase in

creep rate with increasing temperature[2].

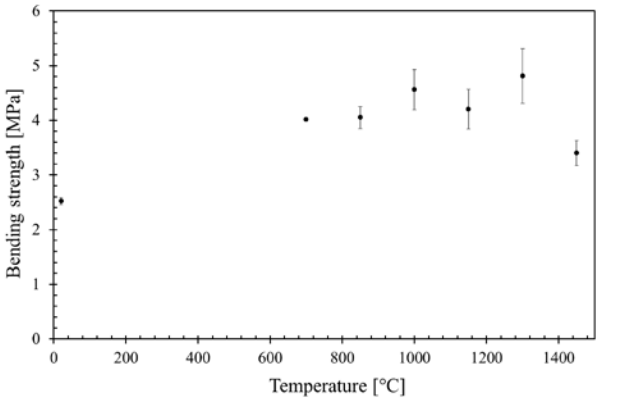


Fig. 5: Bending strength of MgO-C versus testing temperature.

Fracture mechanical tests

The load-displacement curves of the fracture mechanical tests carried out at room temperature, 700°C, 1000°C and 1300°C are shown in Fig. 6. They do not show an unstable crack growth behaviour as would be usual for a wide range of ceramic materials. After a maximum, the load does not fall down abruptly at the beginning of the crack propagation, but shows a rather stable crack growth even at room temperature. It can be seen from the curves that the maximum load increases with increasing test temperature. This means an increasing resistance of the material against crack propagation with increasing temperature. In contrast Hino et al.[9] observed a significant reduction of the maximum load with an increase in the test temperature from room temperature to 1200°C. The reason for this difference might be the higher carbon content of MgO-C used by Hino et al.

Only the load-displacement curve of the experiment at room temperature falls back to zero after reaching the maximum load. All curves from the experiments at high temperatures show a plateau. The load at which the curves remain is higher the higher the test temperature is. The plateau indicates that the crack grows stable until the sample is completely broken.

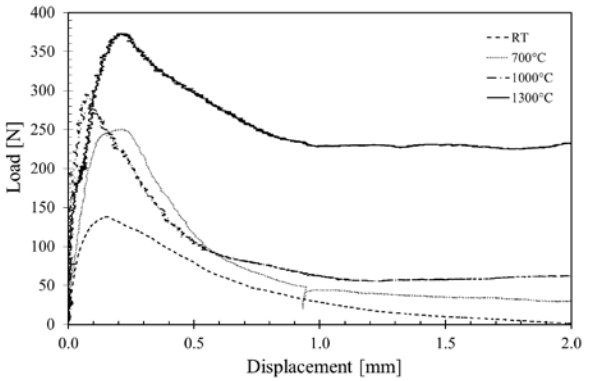


Fig. 6: Load-displacement curve of MgO-C from fracture mechanical tests at different temperatures.

Fig. 7 shows the values of the critical stress intensity factor K_{IC} of MgO-C vs. test temperature. For calculation of K_{IC} the maximum load from the respective load-displacement curves was onset in eq. (3). Thus, the fracture toughness increases with increasing temperature similar to the bending strength. This behaviour can be explained by the closure of cracks and pores in the material with increasing temperature. Additionally, increasing the temperature leads to an increase in ductility of MgO-C, which has a positive effect on the fracture toughness[10].

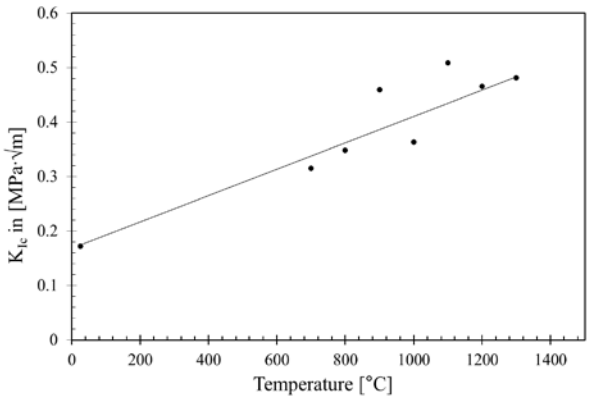


Fig. 7: Development of fracture toughness K_{Ic} of MgO-C with increasing temperature.

Final investigations of the fracture surface by scanning electron microscopy revealed a crack network running through the whole material (see Fig. 8). The cracks often passed the interface of the MgO grains and the matrix, but also through the carbonaceous matrix.

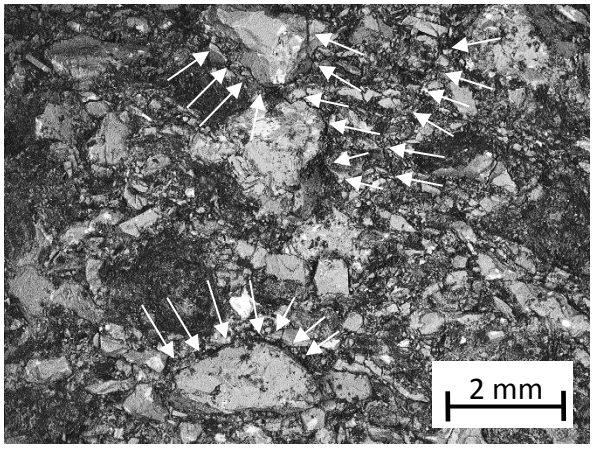


Fig. 8: Fractured surface of carbon-bonded magnesia at room temperature, scanning electron microscopy.

CONCLUSIONS

Bending tests and fracture mechanical tests were performed on MgO-C at room temperature and high temperature up to 1500°C in four point bending tests. The tests were complemented by determination of Poisson´s ratio.

- The material showed a Poisson´s ratio of 0.48 at room temperature.
- The bending strength of MgO-C increases with increasing temperature up to 1300°C, at higher temperature, bending strength decreases.
- MgO-C showed a stable crack growth even at room temperature. The fracture toughness of MgO-C increases with increasing temperature from 0.17 MPa·m$^{1/2}$ at room temperature up to 0.48 MPa·m$^{1/2}$ at 1300°C.
- Investigations of the fracture surface revealed a lot of cracks along the interface between MgO-grains and the carbonaceous matrix.

ACKNOWLEDGEMENT

The authors gratefully acknowledge the German Research Foundation (DFG) for supporting the Priority Programme 1418: Refractory – Initiative to Reduce Emission “FIRE”. In addition, authors would like to thank Mr. Tobias Linemann and Ms. Petra Kästner for sample preparation and Refratechnik Steel GmbH in Düsseldorf (Germany) for supply of magnesia.

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