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Cyclic cold isostatic pressing and improved particle packing of coarse grained oxide ceramics for refractory applications

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ABSTRACT

This study investigated the cold isostatic pressing of coarse grained alumina refractories applying either a cyclic pressure increase or a cycling at maximum pressure. Additionally the effects of the maximum pressure and the particle size distribution on physical, mechanical and thermomechanical properties were analyzed. The cyclic pressure increase resulted in a slightly higher apparent density and lower apparent porosity. A cycling at maximum pressure decreased the median pore size to some extent. Remarkably, an optimized particle size distribution resulted in a lower apparent porosity, lower median pore size and in a higher Young's modulus before and after thermal shock together with a slightly lower relative decrease of the Young's modulus. A higher pressing pressure which decreased the apparent porosity did not affect the Young's modulus. Thus, apparently the optimized particle size distribution improved the particle packing which was associated with a smaller median pore size. This smaller pore size increased the number of pores relative to the total porosity, which then acted as points of crack initiation and crack deflection limiting the length of propagating cracks in case of thermal shock. Thus, tailoring the pore size distribution is a promising starting point to improve the thermomechanical properties of refractories.

1. Introduction

Cold isostatic pressing (CIP) is one of the best-known forming techniques to produce ceramic products with high length to diameter ratios or complicated shapes [1–5]. During cold isostatic pressing according to the wet bag process, powders or granules are filled into an elastic mold, which is later often vacuum sealed. The filled elastic mold is then placed in a fluid and by applying a high pressure on the fluid a very homogeneous compaction of the ceramic body can be achieved [6]. The advantages of cold isostatic pressing compared to uniaxial pressing are much lower density gradients and thus a higher mechanical reliability of the ceramic products [7,8].

Several authors studied the cyclic uniaxial and cyclic isostatic compaction of ceramic, metal and composite powders. Especially a cycling at maximum pressure resulted in smaller density gradients and a stronger densification [9–17].

For single phase powders a considerable higher densification was observed with cyclic pressing, which was explained by an improved particle packing accompanied with a higher particle coordination number and by the fracture of agglomerates [9,14,18,19]. For composites an even higher relative improvement in densification was attributed to the volumetric mismatch between the different phases during a pressure change [11,14,17,20,21].

In general cyclic cold isostatic pressing was only investigated for fine grained ceramics. However, cyclic cold isostatic pressing of coarse grained oxide ceramics such as refractories still remains unstudied.

Thus, the purpose of this study is to describe and examine the effects and possible benefits of cyclic cold isostatic pressing of coarse grained oxide ceramics for refractory applications. Investigated experimental factors were the maximum pressure, the pressure amplitude and the particle size distribution. Furthermore, the difference between a cyclic and linear pressure increase was determined.

2. Experimental

2.1. Materials

The present study investigated the cold isostatic pressing of coarse grained ceramics using alumina as a common refractory raw material. All experiments were performed using full factorial experimental designs.

In all experiments tabular alumina (T60/T64, Almatis GmbH,

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Table 1
Factors of the 2^4 full factorial experimental design. The thermal shock test was included as factor E in the statistical analysis where appropriate resulting in a 2^5 experimental design.

Factor	Identifier	Lower level (− 1)	Higher level (+ 1)
Particle size distribution	A	n_{min} = 0.27 ($Batch_1$)	n_{min} = 0.4 ($Batch_2$)
Maximum pressure	B	75 MPa	120 MPa
Pressure increase	C	linear	cycled
Maximum pressure cycling	D	no (constant)	yes (cycled)
Thermal shock	E	without	with

Germany) was used because it is a widely available raw material [22]. In order to evaluate the effect of the particle size distribution on the cold isostatic pressing, two compositions were designed according to a recently proposed particle size model, which is given in Eq. (1) [23]:

$$CPFT = \left(\frac{d}{d_{max}}\right)^{\left(n_{min} + d \cdot \frac{n_{max} - n_{min}}{d_{max}}\right)} \quad (1)$$

where CPFT is the cumulative percentage of particles finer than a particle diameter d, d_{max} is the maximum particle diameter of the particle size distribution, n_{max} is the distribution modulus at d_{max}, whereas n_{min} is the distribution modulus of an infinitesimal particle size.

The particle size distribution was included as the first factor (A) in the factorial experimental design. The maximum particle size for all particle size distributions was held constant at d_{max} = 3 mm. The particle size distribution was varied by changing the parameter n_{min} according to Eq. (1) on two levels. The parameter n_{max} was held constant at 0.8. The factorial experimental design is summarized in Table 1, whereas the specific composition for each designed particle size distribution is summarized in Table 2.

For all experiments a wax dispersion was applied as a binder (Zusoplast WE 52, Zschimmer & Schwarz GmbH & Co. KG, Germany) because it was previously successfully used to optimize a batch composition for uniaxially die pressed alumina-mullite refractories [24]. The batches were prepared by ordered mixing using a conventional mortar mixer (ToniMIX, Toni Technik Baustoffprüfsysteme GmbH, Germany) [22,24]. In this ordered mixing procedure the coarse grained particle fractions (> 0.5 mm) were first dry mixed for 4 min before the binder was added and stirred for another 4 min. Finally the fine particle fractions were added and the whole mixture was stirred twice for 4 min. Between each step the mixing container was carefully scrapped to ensure a homogeneous mixture. The dry mass of each batch was 3 kg.

The binder amount was optimized for each particle size distribution similar to Schafföner et al. [25]. For this purpose samples with three different binder amounts were uniaxially pressed (ES 270, RUCKS Maschinenbau GmbH, Germany) with a pressure of 120 MPa, see Table 3. The samples were then dried at 80 °C and 110 °C for 6 h at both temperatures before they were fired in air atmosphere with a heating rate of 2 K/min, a maximum temperature of 1650 °C and a dwell time of 4 h. Subsequently, the fired samples were investigated on their apparent porosity and apparent density according to the standard DIN EN 993-1 using water as the immersion fluid. The compositions of each particle size distribution resulting in a low apparent porosity together with an even surface were later used for the cold isostatic pressing experiments.

Table 2
Compositions of the different investigated batches after optimizing the binder amount (see Table 3).

Product name	Raw material	Grain size fraction	$Batch_1$ (n_{min} = 0.27)	$Batch_2$ (n_{min} = 0.4)
Almatis T 60/64	Al_2O_3	1 − 3 mm	40 wt. %	45 wt. %
		0.5 − 1 mm	5 wt. %	10 wt. %
		0 − 0.5 mm	10 wt. %	10 wt. %
		0 − 0.2 mm	20 wt. %	30 wt. %
		0 − 0.02 mm	25 wt. %	5 wt. %
Zusoplast WE 52	PVA binder	mass relative to dry mass	2.50 wt. %	2.50 wt. %

2.2. Cyclic cold isostatic pressing

The samples were pressed in a newly developed cold isostatic press (EPSI N.V., Belgium) with an inner height of 1 m and an inner diameter of 0.25 m which allows a pressure cycling. The used cylindrical molds made of EPDM rubber with a Shore hardness of 40A had an inner effective height of 63 mm after inserting a rubber cap and an inner diameter of 62 mm. To minimize the entrapped air, the filled molds were evacuated for 10 min before pressing.

As a second factor in the factorial experimental design the maximum pressure (B) was varied between 75 MPa (− 1, lower level) and 120 MPa (+ 1, higher level).

To determine the effect of the cyclic cold isostatic pressing, the cycling during the pressure increase (C) and at the maximum pressure (D) were evaluated as two different factors in the factorial experimental design.

At the lower level of the factor C the pressure was linearly increased to the maximum pressure with 1 MPa/s. At the higher level, however, the pressure was cycled during the pressure increase. For a maximum pressure of 75 MPa the pressure was cycled five times during the pressure increase, while for a maximum pressure of 120 MPa the pressure was cycled nine times.

At the lower level of the factor D the pressure was again held constant at the maximum pressure, whereas at the higher level the pressure was cycled for ten times with an amplitude of 25 MPa. The resulting pressure procedures are given in Figs. 1 and 2.

The order of the pressing procedures was random, whereas during each pressure cycle four samples of one of the two compositions were prepared. The order of the two compositions was again randomly chosen for each run of the experimental design.

2.3. Characterization of physical, mechanical and thermomechanical properties

The isostatically pressed samples were dried and fired as the uniaxially pressed ones for the batch optimization. After firing the samples were first examined regarding their apparent porosity and apparent density according to the standard DIN EN 993-1.

Later on, the samples were again dried and a single block with a size of about 20 mm × 20 mm × 20 mm from each batch was cut form the center of the samples using a diamond saw. The cutting ensured an identical sampling for each batch. The cut out blocks were then crushed. Later on, the crushed material was used to determine the pore size distribution using mercury porosimetry (Autopore V 9600, Micrometrics GmbH, Germany) according to the standard DIN 66133.

Furthermore, the thermal shock resistance was evaluated using again a single cylinder of each batch (factor E in the statistical analysis). The thermal shock resistance was determined similar to the standard DIN EN 993-11 using pressurized air. In contrast to the standard, cylinders were used instead of bars. The thermal shock resistance was evaluated by comparing the dynamic Young's modulus before and after a single thermal shock.

The Young's modulus was calculated according to the standard DIN EN 843-2 measuring the velocity of sound. For the calculation of the Young's modulus a Poisson ratio of 0.2 was assumed for alumina, which is consistent with Asmani et al. [26].