Influence of particle size distributions on the density and density gradients in uniaxial compacts

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A R T I C L E   I N F O

A B S T R A C T

This study investigated the influence of particle size distributions on the density, density gradient and strength of uniaxially die-pressed alumina refractories. To design the particle size distributions with a maximum grain size of 1 mm, a modified Andreasen model was applied. It allowed the fine and coarse fractions to be adjusted separately. Cylinders were double-action or two-sided die-pressed. The samples' bulk densities were measured before and after firing (1600 ºC). From the bulk densities, the shrinkage was calculated. The cold crushing strength (CCS) was determined after firing. Some fired samples were axially and radially cut to analyze the density distribution and gradients. Bulk densities before and after firing, shrinkage and strength were highest for increasing amounts of the finest fraction (0–0.02 mm) up to about 55 wt%. Shrinkage and fired bulk density correlated strongly with the CCS. The axial density gradients decreased for increasing fine fraction and decreasing coarse fraction amounts. The radial density gradients were diminished for increasing fine fraction amounts only. Summarized, densest compacts with lowest density gradients were achieved for particle size distributions with a reduced amount of medium sized grains. These batches contained ≈65 wt% fine particles (0–0.2 mm), 10 wt% medium sized grains and ≈25 wt% coarse particles (0.5–1 mm).

1. Introduction

During uniaxial die-pressing of ceramics, density gradients are developed in the compact. Density gradients are density differences over a distance or area. They impact the physical [1,2] and mechanical properties [3] and can cause cracking, warping or distortion after compaction or sintering [4–7]. To improve the quality of uniaxially pressed products, large density gradients have to be avoided.

In a compact, density gradients and the density distribution are caused by the stress distribution induced by the applied compaction pressure [8]. Particularly, the stress distribution develops due to a balance of forces among the external compaction pressure, inter-particle friction and die-wall friction. With the increase of pressure during compaction, firstly inter-particle friction and afterwards die-wall friction occurs [9]. Geometrically, the friction can be expressed as axial and radial friction superposing an internal friction. An increasing angle of the internal friction produces larger stress and density gradients [10–12,2,13,14].

Fig. 1 shows typical density distributions of uniaxially die-pressed compacts. For one-sided pressed compacts, the typical form shows high density regions at the upper edges and in the middle of the bottom [11,13,1] for common height–to–diameter-ratios h/d below ≈1 [15,16].

If two-sided die-pressed, high density regions are in the upper and lower edges as well as in the center of the compact [2].

Generally, the arising density distribution, final mean density and dimensional accuracy depend on various factors. Such factors are the used equipment, like the press mold with the die-play which is the clearance or gap between punches and press mold. The material type, filling behavior and material distribution inside the mold also affect the density distribution. Furthermore, the developing properties depend on operational factors as compaction pressure, compaction speed and the type of de-airing. The final compaction steps like type and speed of pressure relief and de-molding also impact the density distribution and dimensional accuracy [17]. However, although dependences on various factors were identified, only few options to decrease the density gradients were reported in literature.

Different technological measures can improve the density distribution and uniformity of the compact. In general, a decreasing height–to–diameter-ratio h/d leads to smaller gradients [18,2]. Furthermore, a mold roughness with asperities smaller than the particle size reduces the occurring friction [19]. A decreasing pressure and stress lead to decreasing axial and radial friction [20]. Two-sided pressing reduces the density gradients by positioning the neutral plane in the middle of the compact height. If further improved uniformity is required, isostatic compaction can be applied [2,13,17].

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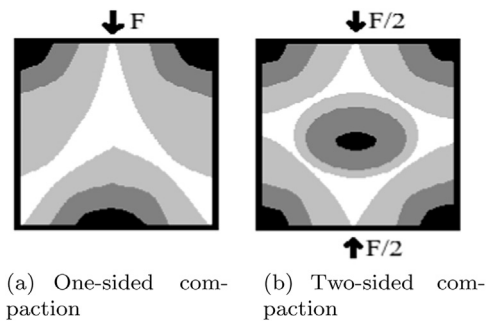(a) One-sided compaction   (b) Two-sided compaction

Fig. 1. Typical density distributions of uniaxially die-pressed compacts (higher density regions are darker) [2].

Additionally, the compact uniformity is influenced by the material and batch type as well as the batch composition. Reducing the content of fines in the material leads to decreasing friction [19]. It was also suggested that the application of less irregularly shaped particles reduces friction [19]. Additionally, lubricants or pressing aids are commonly applied to improve the pressure transmission and to reduce the density gradients [2,14]. The lubricant has to be chosen according to the surface chemistry of the particles [21]. For example, humidity is reported to increase the density gradients in alumina samples [18,22]. Besides internal lubricants, also die-wall lubrication reduces die-wall friction and the developing density gradients [11]. However, mixing a lubricant with the powder is more efficient concerning the reduction of friction than die-wall lubrication [3]. Furthermore, an increasing packing density increases the uniformity of the compact [6]. In a densest packing there are numerous inter-particle contact areas or points rather than individual contact points [22]. The number of contact points per particle, the particle coordination, increases with increasing packing density [23]. Consequently, in a densest packing the pressure is rather transmitted than dispersed which improves the compact uniformity [22].

However, all the aforementioned options were reported only for powders or granules with maximum sizes up to 300 μm. To the authors' knowledge there were no studies for coarse grained ceramics like refractories. In a previous study [24], the effect of different lubricating additives on the compaction of constant compositions with a maximum grain size of 6 mm was investigated. It was found that the density gradients were statistically not influenced by the application of the additives. It was suggested that the particle size distribution plays a key role for the developing density distribution in coarse grained ceramics.

The purpose of this study, hence, is to investigate the fundamental dependence of the density gradients on particle size distributions containing also coarse grains. Therefore, densities and density distributions of two-sided die-pressed alumina samples with a maximum grain size of 1 mm were measured. These properties were furthermore related to the cold crushing strengths of the samples.

The fundamental results will be applied to press large crucibles with reduced porosity and other parts for the steel casting simulator [25].

2. Material and methods

The current study involved designing particle size distributions and measuring density, density distribution, shrinkage, and cold crushing strength (CCS) of pressed cylindrical samples.

The raw materials were four alumina fractions (Tabular Alumina T60/64, Almatis GmbH, Germany) up to 1 mm (cf. Table 1) and a pressing aid (Zusoplast WE 52, Zschimmer & Schwarz GmbH & Co. KG, Germany), similar to Fruhstorfer et al. [26,27]. The particle size distributions and true densities of the single alumina fractions were measured previously [26]. They are required to calculate the single fractions' amounts for the batches according to a modified Andreasen model, also introduced in the previous study [26].

Table 1

Compositions of the pressing masses in wt% with adjusted minimum distribution modulus n_{min} and maximum distribution modulus n_{max}.

n_{max}	Fraction	n_{min}			
	in mm	0.2	0.4	0.6	0.8
0.4	0.5–1	20	15	25	25
	0–0.5	10	40	40	55
	0–0.2	15	25	35	20
	0–0.02	55	20	0	0
0.6	0.5–1	25	25	30	30
	0–0.5	10	30	35	50
	0–0.2	10	25	35	20
	0–0.02	55	20	0	0
0.8	0.5–1	30	30	35	35
	0–0.5	10	30	35	45
	0–0.2	5	20	30	20
	0–0.02	55	20	0	0
1.0	0.5–1	35	35	40	40
	0–0.5	10	25	35	45
	0–0.2	0	20	30	15
	0–0.02	55	20	0	0

The modified Andreasen model $A_V(d)$ (Eq. (1)) describes the cumulative percent finer than the particle diameter d. The standard Andreasen model [28] applied a constant distribution modulus n which is independent of d. In contrast, the new approach applies a linear modulus function $n(d)$ (Eq. (2)) dependent on the particle size. Therefore, the amounts of fine and coarse particles can be adjusted separately by the minimum and maximum distribution modulus n_{min} and n_{max}, respectively. In the present study, n_{min} was adjusted on the levels 0.2, 0.4, 0.6 and 0.8 as well as n_{max} on the levels 0.4, 0.6, 0.8 and 1.0. The maximum particle size d_{max} was kept constant at 1 mm. The resulting compositions are presented in Table 1. In all batches, 2.5 wt% pressing aid was added.

$$A_V(d) = 100\% \cdot \left(\frac{d}{d_{max}} \right)^{n(d)}$$
(1)

$$n(d) = n_{min} + d \cdot \frac{n_{max} - n_{min}}{d_{max}}$$
(2)

The pressing masses of 2.5 kg batch size were prepared with a concrete mixer (ToniMIX, Toni Baustoffprüfsysteme GmbH, Germany). Firstly, the coarsest fraction was mixed with the liquid pressing aid for 1 min. Subsequently, the finer grain fractions were added and it was mixed for 3 min. After scratching the mixer walls, the batch was mixed for 3 more minutes. This mixing procedure is referred to as ordered mixing [24,29,30] and results in fine particles adhering to the coarse grains.

Six cylinders with a diameter of 50 mm were then double-action, respectively two-sided, die-pressed with a hydraulic press (ES 270, RUCKS Maschinenbau GmbH, Germany). In fact, pseudo double-action pressing was applied which means that the lower punch was fixed but the die-matrix moved downwards with half the speed of the upper punch. The filling height was 80 mm. It was twice de-aired at 1/3 and 2/3 of the maximum pressure of 50 MPa. This pressure was chosen to avoid lamination cracks in the batches with high amounts of fines. After ejection, the diameters and heights of the cylinders were measured three times on different locations. Furthermore, the samples were weighed and then their unfired bulk densities calculated.

Afterwards, the cylinders were sintered in an electric chamber furnace (HT 16/18, Nabertherm GmbH, Germany). They were heated with a heating rate of 1 K/min up to 600 °C and then with 2 K/min to the sintering temperature of 1600 °C. The sintering temperature was held for 1 h before the furnace was turned off and the samples cooled freely inside. Then, the samples' dimensions, weights and thus fired bulk densities were determined with the same methodology as before sintering.