SOME CONSIDERATIONS REGARDING THE GRAIN SHAPE OF REFRACTORY RAW MATERIALS

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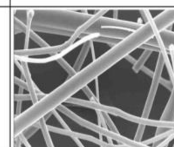
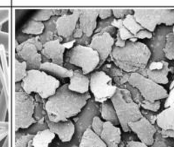
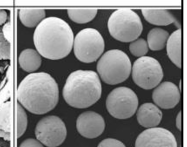
ABSTRACT
The particle size distribution (PSD) is an essential characteristic of raw materials. This feature is taken into account by nearly all aspects related to the development and optimisation of refractory products. Nevertheless, not only the PSD itself is important but also the shape of the grains. This has long been neglected especially within the field of refractories. For e.g. building materials, road construction and abrasives it is common practice to characterise raw materials not only regarding their size distribution but also concerning their shape.
Like the particle size distribution also the grain shape is already determining several important properties of the raw material itself as well as of their mixtures. This might be reflected in the behaviour of a material discharged from a silo, the miscibility of different raw materials, the flowability of a readily mixed composition into a mould, the compressibility etc. Moreover, the properties of refractory products such as porosity, strength, abrasion, corrosion as well as creep resistance might be significantly influenced not only by the size distribution but also by the shape of the raw material particles. Finally it is well known that the reactivity of a particle is determined by its specific surface area and this again is depending on the shape.
Therefore in a first part of this work some general considerations regarding the definition of grain shape, its measurement and the targeted production of raw materials with certain grain shapes will be presented. A second part is then dedicated to the influence of differently shaped raw materials on the properties of refractory castables and bricks. As a result it is shown that raw materials with identical size distribution but with differently shaped grains can influence the physical, mechanical and thermo-mechanical properties of refractories.

INTRODUCTION
Refractory raw materials are usually comprehensively characterised regarding their chemical and mineralogical composition, their physical properties like bulk density and porosity and their size distribution. The latter is of special interest in order to achieve a dense packing and good flow properties in case of castables. Nevertheless, another important characteristic feature of the grains / aggregates is mostly not considered – their shape, even though this is often crucial for the intended application of the final product.
Shape is generally meaning the particular physical form or appearance of something, its external outline or surface and the terms morphology, habitus or form are often used as synonyms. The simplest differentiation regarding shape is the number of dimensions of an object. With respect to the macroscopic world a fibre would represent an one-dimensional, a platelet an two-dimensional and a sphere an three-dimensional object. Examples of refractory raw materials (natural and artificial) exhibiting these shapes are given in tab. 1 together with dedicated applications.
However, most of the refractory raw materials have to be considered as three dimensional objects but they don't necessarily correspond to a sphere. Therefore further differentiations are required in order to describe the shape more precisely.
Besides the shape itself, also the roundness of the grains and their surface texture might be of interest.

Tab. 1: Shape of refractory raw materials in dependence on the three spatial dimensions

Shape types:	Fibre	Plate	Sphere
Dimensions:	1	2	3
SEM picture:			
Example			
Natural:	Asbestos	Clay	Beach sand
Artificial:	Alumina Fiber	Boron Nitride (hex.)	Bubble Alumina
Application:	Insulation	Coating	Sand casting

Shape measurement
A simple way to analyse the shape of an individual particle is the comparison with shape charts and indices. Several different have been developed e.g. by Zingg [1], Krumbein [2] and Powers [3]. Nevertheless, a statistical verification is laborious and time consuming.
Nowadays powerful camera or laser based systems are available enabling the automatic analysis of a large number of particles.
As parameter for the characterisation of the shape often either the width / length ratio:

$$\frac{b}{l} = \frac{x_{c\,min}}{x_{Fe\,max}}$$

$x_{c\,min}$ shortest chord of a particle projection
$x_{Fe\,max}$ longest direct length of a particle projection

or the sphericity:

$$\mathrm{SPHT} = \frac{4\pi A}{U^2}$$

A: measured area of a particle projection
U: measured circumference of a particle projection

is used. For both parameters a fibre would give a value approaching to zero whereas in case of a perfect sphere the rate will be 1. Thus, it is possible to characterise refractory raw materials regarding their shape and like for the grain size one can also record a grain shape distribution.
Doing this for different products, it soon becomes apparent that the shape distribution sometimes varies significantly and the question arises, how is the shape of our raw materials influenced.

Production of different shapes
Besides the direct extraction of naturally shaped minerals like asbestos, clay, graphite, talk or sand, there are different possibilities to influence the grain shape of a raw material. Principally one can differentiate between a direct production, a targeted processing (comminution / treatment) or a shape separation. Tab. 2 shows an overview about the different possibilities.
However, it needs to be stated that most of the technological processes usually lead to a more or less broad range of different grain shapes. In fig. 1 an example is show for the influence of the type of crusher on the sphericity of a corundum raw material.
It can be seen from fig. 1 that the use of different crushers is significantly influencing the grain shape. By using the roller crusher a clearly more angular grain is produced whereas the processing

with an impact crusher results in a comparatively blocky grain. Nevertheless, even the same type of crusher not necessarily leads to the same grain shape distribution as one can see from the second corundum in fig. 1. The material itself (e.g. the sintering degree) as well as some technological variations of the used equipment has an impact on the resulting shape distribution too.

Tab. 2: Production of differently shaped raw materials

Method	Process	Shape
Direct production	Melting + Atomising	Fibres, Spheres
	Extrusion	Rods, Platelets
	Tape casting	Platelets
	Agglomeration / granulation	Spheres
	Spray drying	Spheres
	Spray pyrolysis	Spheres
Comminution / Treatment	Roller crusher / mill	Angular grains
	Cone crusher	Angular grains
	Impact crusher / mill	Blocky grains
	Ball mill	Blocky grains
	Flash calciner	Spherical particles
Shape Separation	Sieving	
	Friction sorting	
	Optical sorting	

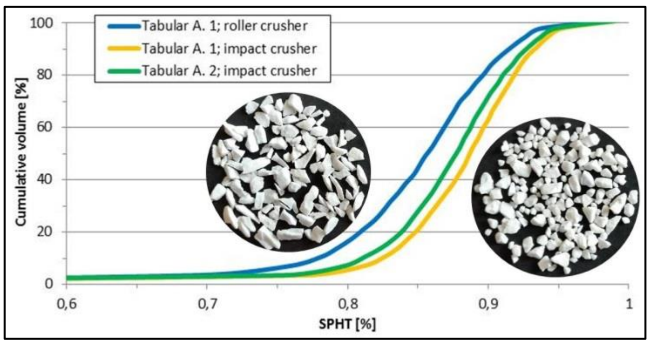


Fig. 1: Dependence of the grain shape on the crusher technology

For our investigations we used partly Tabular Alumina and also WFA from which we sorted for each of the size fractions 0,1-0,5mm, 0,5-1mm, 1-3mm and 3-6mm an either blocky or angular material by using a type of Wilfley table [4]. Since such a vibration table is not only sensitive regarding the grain shape but also concerning the size of the particles, especially care was taken in order to ensure an identical particle size distribution for the different shape fractions.

INFLUENCES ON REFRACTORY PRODUCTS

In the following some results will be given regarding the influence of a different grain shape on the properties of refractory products (castables and bricks). In all cases a careful adaption of the grain size distribution was necessary due to the variations in the apparent density in order to ensure identical volume fractions within the final product.

The impact of the grain shape was investigated for the three areas:
- workability of refractories;
- physical properties of refractories;
- thermal properties of refractories.

Workability of refractories

It is known that, besides the particle size distribution, also the particle shape effects the flowability of dry powders as it is e.g. described for the behaviour of soil [5]. Thus, the filling of a mould for dry pressing might already be influenced by the grain shape. Even more interesting are the flow properties of castable or gunning mixes. As it has to be expected and is already described in literature [6, 7, 8] spherical particles used either as ultrafines (e.g. microsilica) within the matrix or as coarser aggregates will lead to a better flow characteristic of monolithics. However, especially the aggregates are often not really spherical but resulting from a crushing / milling process with a more blocky or angular shape as it is shown in fig. 1.

In a first attempt we used the obtained WFA materials to investigate the influence of the grain shape on the flowability of a vibration and a self-flowing castable. The results can be seen from fig. 2.

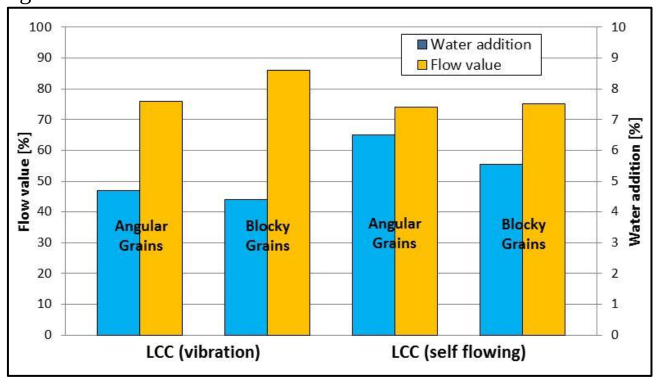


Fig. 2: Water addition and flow values for different castables

As one can see, a more blocky grain shape of the raw material leads to either a better flow value by identical water amount or to a reduced water demand for a comparable flow.

The either angular or blocky shaped grain fractions of the WFA were additionally used to prepare dry-pressed samples (cylinder of 50mm height and 50mm diameter or prisms of 25*25*120mm). Also in this case a precise adaption of the grain size distribution was ensured and the mixing and pressing procedure was fully identical. Nevertheless already the green density was with 3,32g/cm^3 higher for the bricks on base of the blocky grains in comparison to the angular grains (3,27g/cm^3). A higher density of refractories due to more spherical aggregates is already mentioned in literature [9]. Obviously the mixture containing the angular grains is more difficult to compact as the blend on base of the blocky aggregates. This is likely in connection with the higher surface of the angular particles and thus, with the higher friction during the compaction of this mix.

Another effect which should additionally be considered by working with elongated or platy particles is their possible orientation within the final product. In dependence on the shaping technology such particles tend to a preferred alignment perpendicular to the compression direction. Such effects can be observed for the fibres in vacuum formed boards, for dry-pressed and extruded bricks as well as for castables. Fig. 3 shows an example for dry-pressed bricks containing the angular WFA aggregates.

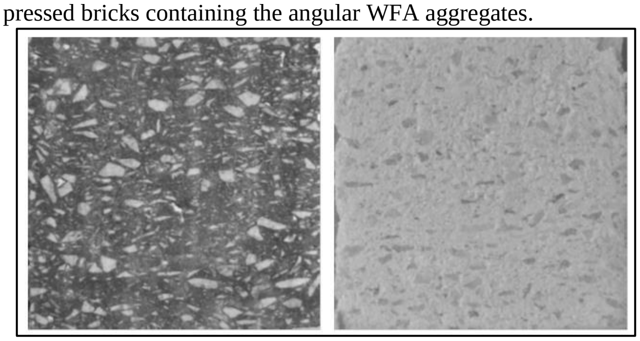


Fig. 3: Preferred horizontal orientation of angular corundum aggregates in the dry-pressed WFA bricks

Physical properties

It is clear that the mentioned different water demand of the castables (fig. 3) in dependence on the grain shape will lead to some variations regarding the density, porosity, permeability and the mechanical properties too.

Differences of the physical properties in dependence on the grain shape could also be found for the WFA bricks after drying and sintering (1750°C / 5hrs). The results of their characterisation regarding shrinkage, density and porosity are summarised in tab. 3.

Tab. 3: Physical characterisation of dry-pressed bricks

		WFA Blocky	WFA Angular
Geometric Determination:			
Density	[g/cm^3]	3,35 ±0,01	3,30 ±0,01
ΔL	[%]	-0,49 ±0,09	-0,71 ±0,05
ΔV	[%]	-1,64 ±0,15	-1,70 ±0,07
Water Adsorption:			
Density	[g/cm^3]	3,35 ±0,01	3,26 ±0,01
Open Porosity	[%]	14,9 ±0,23	16,3 ±0,21
Hg-Porosimetry:			
Density	[g/cm^3]	3,32	3,26
Open Porosity	[%]	15,4	17,4

Even by ensuring the same grain size distribution and identical pressing procedure as well as sintering conditions the blocky grains generally lead to a higher density and lower porosity of the bricks. It is interesting to note from the data in tab. 3 that the linear shrinkage is higher for the angular grains even the volume change is comparable to the bricks on base of blocky WFA. This is obviously a clear hint on the above mentioned problem with the orientation of the angular aggregates after pressing (fig. 3). The higher amount of fines in the direction of the compaction is causing the higher shrinkage whereas perpendicular to this the amount of fines is lower and thus the shrinkage is not as pronounced. For the whole brick these two effects within the different directions compensate each other and the volume change is nearly identical for both types of aggregates.

This orientation also affects the mechanical properties of the dry-pressed bricks. In fig. 4 the Youngs Modulus determined for the different sample directions is plotted. One can see that in the vertical direction (pressing direction) the values determined for the angular aggregate containing bricks are lower than for the product on base of the blocky grains whereas no significant effect could be found in the horizontal directions. This direction depending difference is most likely caused by the preferred orientation of the angular grains during compaction.

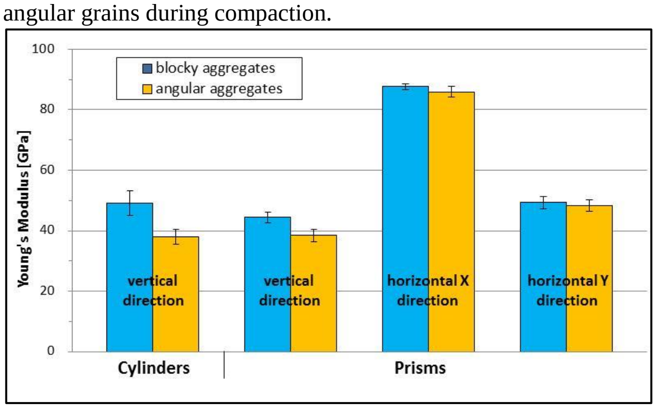


Fig. 4: Youngs Modulus for the dry-pressed bricks in dependence on the orientation

A similar effect obviously caused by an orientation was observed for the elastic properties of the WFA castables after drying and pre-sintering at 1500°C for 5hrs. Fig. 5 shows the Youngs Modulus of the different castables measured in either vertical or horizontal direction.

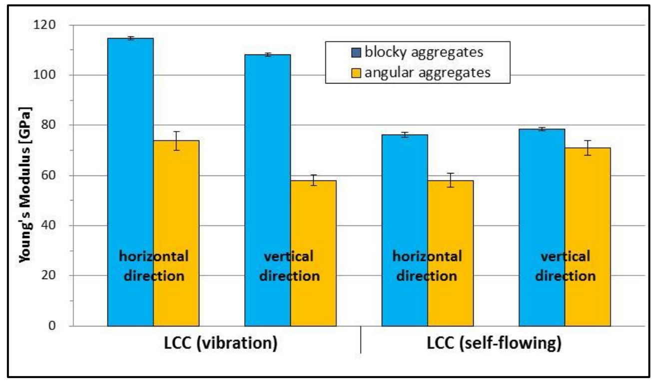


Fig. 5: Youngs Modulus for the castables in dependence on the orientation (pre-sintered at 1500°C)

It becomes obvious that also in this case the two castables on base of blocky grains exhibit a significantly higher Youngs Modulus than those on base of the angular grains. However, there is another interesting result and this is connected with the already mentioned preferred orientation of the angular grains. As one can see from fig. 5 there is no significant difference in the elastic properties of both castables on base of blocky grains in dependence on the measurement direction. Nevertheless, for the angular grains there is obviously a direction-depending effect. Surprisingly the trend is different for the vibration and self-flowing castable. In the first case the Youngs Modulus is lower in the vertical direction whereas for the self-flowing mixture this value is significantly higher than that for the horizontal direction. This result is probably caused by a different orientation of the grains within these two castables.

The results obtained for the bending strength of the different castables followed the same trends as the Youngs Modulus. The higher strength for the castable containing blocky aggregates is due to the denser packing and thus, in line with results from literature [10].

High temperature properties

With the bricks on base of the two different grain shapes we did thermal shock tests and creep measurements in order to investigate the influence on the thermo-mechanical properties.

The results of the thermal shock tests are summarised in fig. 6.

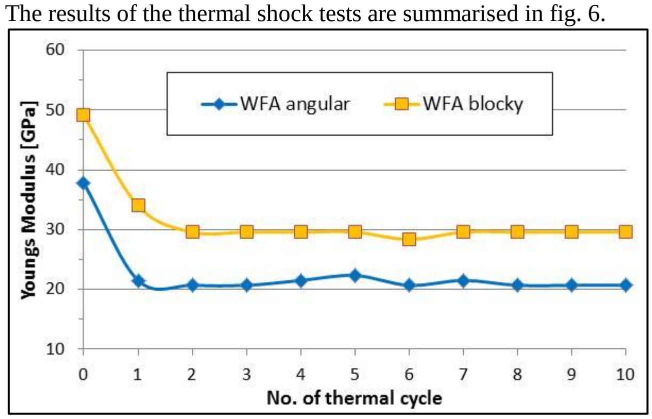


Fig. 6: Thermal shock resistance of WFA bricks

The general course of both samples is identical but, as one can see from fig. 6, the blocky WFA obviously leads to a higher Youngs Modulus of the bricks than the angular material.

Also creep measurements were carried out at 1680°C. The creep process during 25hrs is plotted in fig. 7.

As already for the thermal shock resistance, the blocky aggregates are also leading to clearly better results for the creep resistance (lower creep rate) than the angular grains.

Thus, it seems like blocky grains are positively influencing the thermo-mechanical properties of refractory bricks. However, from the results in tab. 3 it becomes clear that the blocky grains are causing a higher density / lower porosity of the bricks compared to the angular material. Since both - thermal shock as well as creep resistance - are significantly influenced by density and porosity of the material, it is not possible to conclude on a direct impact of the grain shape.

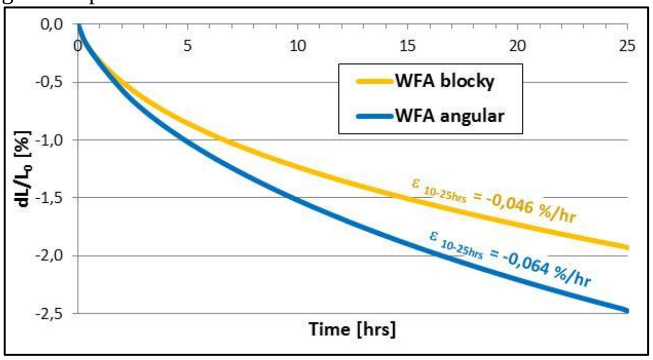


Fig. 7: CiC of WFA bricks at 1680°C

With the pre-sintered samples (5hrs at 1500°C) of the vibration and self-flowing castables we did also creep measurements at 1400°C. For this we drilled cylindrical samples from bigger casted blocks in either horizontal or vertical direction. Fig. 8 shows the course of the creep in compression for all materials. Additionally the creep rate between 10 and 25hrs is separately plotted in fig. 9.

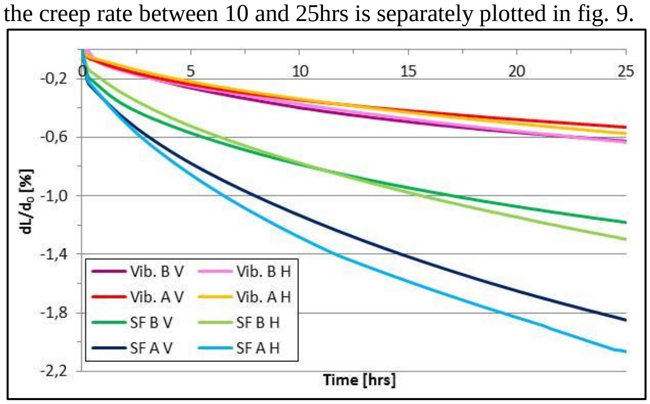


Fig. 8: CiC for different castables (Vib.–vibration castable, SF–self-flowing castable; B – blocky, A-angular; V-vertically, H-horizontally oriented)

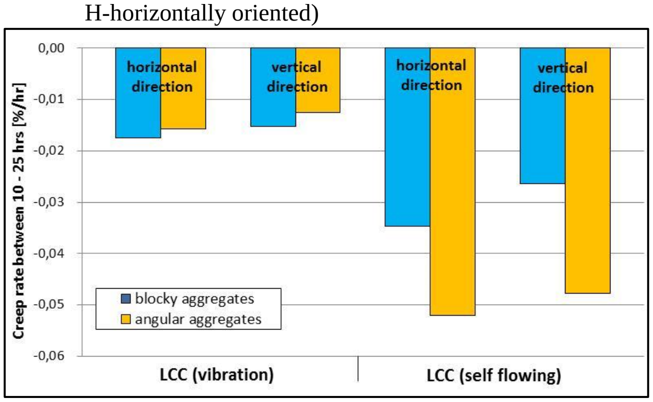


Fig. 9: Creep rate of different castables between 10 and 25hrs

The self-flowing castable generally exhibits a higher creep in comparison to the vibration castable. This is caused by the higher amount of fines and in connection with this the higher water demand necessary to achieve the self-flowing effect.

The creep rate is always slightly higher for the samples which were drilled from the horizontal direction of the casted blocks and even this effect is rather small, it occurred for all of the different materials.

Also the grain shape seems to influence the creep rate of the castables. In case of the vibration castable no clear trend can be seen indeed, but for the self-flowing mixture the angular aggregates are obviously causing a significantly higher creep, independent on the sample direction. However, this might also be caused by the higher water addition (see fig. 2) and consequently by the higher porosity and lower density.

CONCLUSION

In dependence on the crushing / milling technology refractory raw materials usually contain differently shaped grains in varying quantities. Despite of this, up to now refractory producer are mainly focused on the size but not on the shape of the used feedstocks.

For some fundamental investigations on the influence of the grain shape on workability, physical and thermo-mechanical properties of refractory castables and bricks, we separated either blocky or angular material from a standard WFA by using a vibration table.

It could be shown that the properties of the refractory products are significantly influenced by the grain shape. The main differences found are obviously either related to the workability (compressibility, flowability) or to the alignment of the grains in a preferred direction. Due to these effects we found for both, castables and bricks, in case of the blocky grains usually higher density / lower porosity, better mechanical properties, a higher thermal shock and a better creep resistance. Thus, the shape of refractory raw materials is an important feature which should always be considered.

The preferred orientation of elongated grains in dependence on the type of shaping technology for the refractory products might be a future possibility for the intended design of certain anisotropic characteristics like e.g. a lower rebound of gunning mixes, higher corrosion-, thermal shock – or abrasion resistance in load direction.

Finally it needs to be said that besides the shape there are two other factors which are influencing the behaviour of our raw materials, the angularity or roundness and the surface texture.

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