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Deep Drawing of Ceramic Green Tapes – A Promising Technology for Producing New Lightweight Non-Planar Kiln Furniture

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Abstract

For the processing of ceramic green tapes to form 3D components, different technologies are known, for instance lamination to multilayer structures or folding. In the metals and plastics industries, deep drawing is, in addition to bending, a widely known method for reshaping sheet metals and polymers. This technology is not, however, applied for processing ceramic green tapes because of the downstream debinding and sintering steps. During the complex shaping process, the ceramic particles arranged within the polymer matrix become rearranged, which can result in inhomogeneous particle distribution. During sintering, this can lead to defects because of the resulting inhomogeneous shrinkage behavior. This study presents basic investigations into the deep drawing of ceramic green tapes. A special method was developed to investigate the particle distribution within the green tapes before and after the deep drawing and to correlate the results with the process conditions and the location of the investigated volume. One possible application for deep-drawn ceramic tapes could be as pre-product for non-planar kiln furniture. In combination with extruded pre-products, lamination, and co-sintering, large components with a low density and thermal mass can be produced with a top surface geometry similar to that of the sintered products.

Keywords: Structural ceramic, tape casting, shaping, forming, deep drawing

I. Introduction

Tape casting in the scope of multilayer technology is a well-known process for producing wide and flat two-dimensional components known as preceramic green tape, with thicknesses in a range of 5 to 2000 µm. The process is ecological providing a water-based system [1–3] is used. The green tapes can be processed, for example by lamination to multilayers, by stamping or by folding. Deep drawing – a new method for processing green tapes, which was originally developed for the metals and plastic industries [4–6] – shows high potential for the realization of complex-shaped ceramic structures in a relatively easy process, which can then be used e.g. to make housings or to produce new lightweight kiln furniture, especially with a non-planar surface.

The widely applied process of deep drawing was also investigated for technical ceramics [7]. Applying the technology for making technical ceramics is one possibility to increase the complexity of planar ceramic green tapes. Schuhmacher *et al.* have shown the possibility of deep drawing ceramic green tapes in principle.

In general, kiln furniture has to ensure the safe support or transport of ceramic products during thermal processing. Therefore, and especially at high temperatures, compressive strength and high bending strength are needed. Thick, dense or porous ceramic plates are state of the art, but the high thermal mass of these structures has to be heated. A homogeneous temperature distribution can only be achieved at low heating or cooling rates. Higher rates result in thermally induced mechanical stresses, which can damage the kiln furniture and the ceramic products.

To increase the possible heating and cooling rates as well as to reduce the energy consumed for heating up the kiln furniture, a new generation of these products is needed. Several papers show alternative structures with macroscopic pores and open cavities allowing the passage of gas or air through them [8–10].

Further works undertaken by our group showed another technology to produce planar lightweight structures for kiln furniture with following properties [11–14]:

- low thermal mass and heat capacity
- open macroscopic cavities allowing the passage of air
- high stiffness and strength at elevated temperatures
- high thermal shock resistance

Innovative kiln furniture based on sandwich-like structures with laminated green tapes as top and bottom surfaces were realized with non-shaped and shaped green tapes or extruded components with different geometries.

Different papers presented this new type of kiln furniture and the technologies for its production [11–13], the stiffness of its structure at high temperatures and when supporting products [12], a high time and energy saving potential (up to 40 % respectively) [13,14], technologies for the mass production of these structures [14], and a significant improvement of the temperature distribution within the

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kiln [14]. Another paper [15] described basic research on the deep drawing of ceramic green tapes as a function of different properties, e.g. green tape composition, stamp geometry and drawn depth. The general idea is to combine reshaped green tapes with a supporting structure that has been extruded and machined, as shown schematically in Fig. 1.

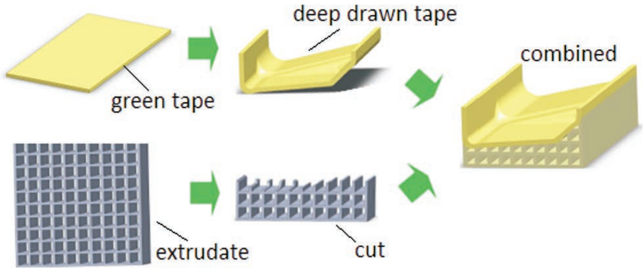

Fig. 1: Idea of combining a reshaped ceramic structure with a ceramic extrudate as support.

Different crucially important challenges must be solved for realization of the approach described above. One challenge is joining the different components in the green state, to ensure a high-quality interface after sintering. This was realized for planar structures in former work [13]. Another challenge is the re-shaping of the green tape, necessitating the adaption of the deep-drawing process for ceramic green tapes. Deep drawing of ceramic green tapes without a subsequent thermal process is possible, but the debinding and sintering can be challenging depending on the degree of reshaping. During deep drawing of green tapes, the particle-polymer distribution relates to local changes depending on different factors, e.g. degree of drawing, binder/polymeric system, powder content and tool friction, which all need to be investigated more thoroughly.

(1) Deep drawing – a new way to form ceramic green tapes, achieving higher complexity

In accordance with DIN 8580, reshaping is classified in the second group of manufacturing technology and means a change in the plastic status of a solid structure. Alongside bending, deep drawing is one of best-known methods for reshaping sheet metals. It is mainly used in the metals and plastics industries. This technology is used to cost-efficiently reshape semi-finished products such as flat sheets and tapes to more complex 3D components. Original products are, for example, texture components used in the automotive industry, dishwasher and washer components in white goods for domestic and commercial use, emesis basins in the field of medical engineering, and receptacles for food. Although there are different deep drawing categories and the most commonly applied method is deep drawing with tools. Thereby, an open hollow body is produced in a so-called first draw. The schematic in Fig. 2 shows the deep drawing process, before (left) and after drawing (right) of a green tape. During the process, a sample is fixed to a matrix by a hold-down clamp, normally with a pre-defined pressure. Next, the stamp moves down into the sample (z-direction), force- or path-controlled, drawing the sheet into the matrix to form a hollow by compressing and stretching.

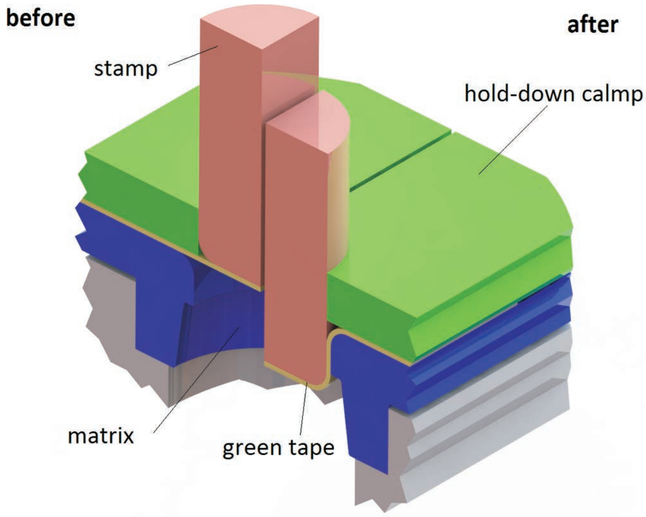

Fig. 2: Schemata showing the deep drawing process before (1) and after drawing (2) a green tape.

Depending on their structure, metals and plastics obtain their final form after the forming process. The products can be used and handled immediately after drawing, for example, for the application of certain a finish with paint, etc. However, deep drawing of ceramic green tapes is not state of the art and needs to be researched more thoroughly. Green tapes are an intermediate form of the final ceramic structure. After the green tapes have been cast, they can be shaped in a follow-up process, but then they have to be processed in the conventional ceramic way involving debinding and sintering. That is a challenge because schematically the green tape is a polymer, high-filled (up to 67 vol%) with ceramic particles. Strictly, it can be considered as a particle-disturbed polymer.

The focus of this work is the investigation of deep-drawn ceramic green tapes and the development of an examination method to provide a basis for developing a technology for the production of the new lightweight kiln furniture with a non-planar surface described above.

II. Experimental

During the study, firstly the relationship between shaping behavior, green tape properties and forming parameters was investigated. Therefore, green tapes based on alumina (A16 SG, Almatis) particles with a solid content of 40 to 60 vol% were realized with variations of binder systems, especially the ratio of binder and plasticizer, and three different green tape thicknesses were realized. Binder for strength and plasticizer for flexibility define the general properties of a green tape at a constant particle content. Mixtures of polyvinyl alcohol (PVA) as a common binder [16] and glycerin as plasticizer, both with a content of 10 to 20 vol%, were used as the binder system. PVA is a thermoplastic polymer with a glass transition point of 85 °C. With the addition of a plasticizer, this point could be moved to lower temperatures, softening the polymer. Therefore, the flexibility of the ceramic green tape, especially of its forming properties, depends on the ratio of binder to plasticizer.

By way of example, the preparation of a suspension for a green tape with a solid content of 60 vol% is described.

For its preparation, the alumina powder was deagglomerated in an aqueous solution with a dispersant and defoamer in a planetary ball mill for one hour. After this deagglomeration, the particles were homogeneously dispersed and formed a suspension with a solids content of about 80 mass%. After addition of binder, plasticizer, surfactant and defoamer, the suspension was homogenized for 12 h on a roller chair. After this treatment, the suspension had a solids content of about 50 vol% and a pH of 9. The suspension was de-aired by gently stirring under vacuum before casting.

For the tape-casting experiments, laboratory equipment was used. This discontinuous tape casting machine has a moveable doctor blade and the suspension is cast upon a stationary polymer carrier. During the drying process, it is possible to regulate temperature and humidity of the air flow over the wet film of the suspension. Thereby the risk of skin formation and cracking can be reduced, because the drying air has been saturated with solvent vapor when it meets the wet film of suspension. Casting gap width was varied in a range of 1.1 to 2.0 mm to generate tapes with a thickness of 300 μm and 600 μm.

The preparation of an alumina suspension as well as the casting and lamination process have been described in more detail in other papers [17–19].

In the next step, a deep-drawing test unit based on a linear axis with spindle drive, which can be controlled in one direction with a resolution of 0.01 mm, was developed. Before starting the drawing tests, the tapes were marked with a grid pattern by stamping with conventional ink. Fig. 3 shows the developed deep-drawing unit, the grid pattern and a green tape with a grid fixed within the test unit.

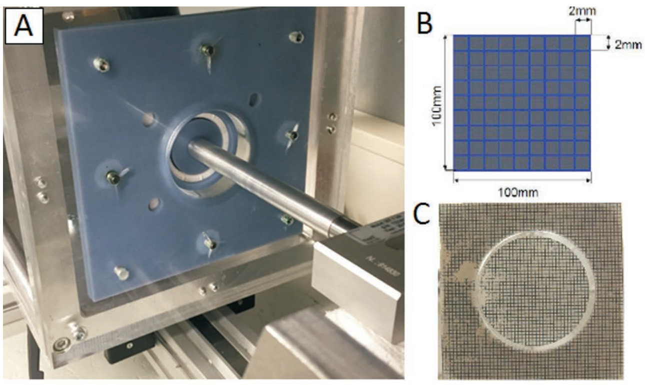

Fig. 3: Developed test unit for basic investigation of the deep drawing of ceramic green tapes (A) grid pattern with mesh size of 2 x 2 mm (B) used to grid the green tapes (C).

The deep-drawing unit consist of two slides, one active and one passive. The passive one is manually controlled and moveable over the complete axis. On this slide, the supporting structure for the matrix was installed. The active slide is directly controlled by the axis. The mount for the stamp and the load cell (10000 N ± 0.2 N) is attached on the top. With the spindle drive, the second slide can be moved precisely to direct the stamp into the matrix fitted with the ceramic green tape to be deep drawn. The load cell was used to measure the forces during drawing, in order to correlate them with the properties of the green tapes. For the first test series, a relatively simple stamp geometry that could be easy calculated was deemed a useful tool. We, therefore, performed the first test with simple machined half-spherical stamps in diameters of 20, 40, and 60 mm consisting of aluminum with a smooth surface. The stamp induced the form of the shaped sample and applied the necessary force for forming. With the grid pattern, the deformation of the green tapes during drawing was visualized and the change in the grid squares after the drawing could be characterized.

After deep drawing of the green tapes, the measured forces were characterized as a function of the parameters used. In the next stage, the strain state of the green tape was characterized. For this purpose, an optically based 3D-scan of the shaped green tapes and the grid squares was performed and evaluated. It was necessary to develop a special methodology to characterize the structure of the ceramic green tape before debinding and sintering.

The approach was to determine the structural state of the shaped green part, especially the density distribution in the different areas and the distribution of the powder particle. Because of sintering shrinkage and possible geometric delay, the completely sintered structure does not allow any conclusions regarding the deformation state in the green part. That means the state of the shaped green tape structure must not be changed. Based on this, the formed tapes were debinded and sintered at a maximum temperature of 900 °C without dwell. Shrinkage within the structure could be virtually avoided, and adequate strength enabling handling of the component after thermal treatment was achieved because the first sinter necks were formed between the particles. To visualize the inner structure and to get topographical information about the particles, analytical techniques such as microscopy were used. The structure was characterized with microscopy, especially FESEM (Field Emission Scanning Electron Microscopy). Therefore, a longitudinally cut component was necessary. To obtain this, the component was embedded in epoxy resin and sliced over a cross-section.

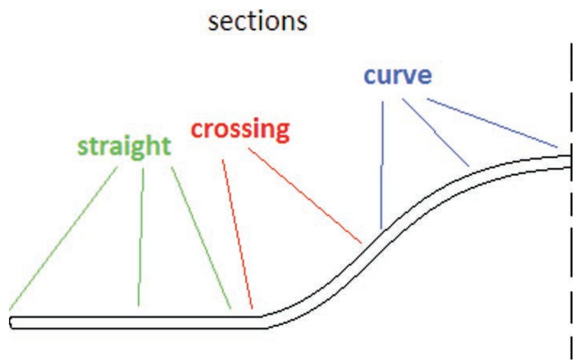

Fig. 4: Virtual division of the embedded deep-drawn component into different sections.

Having been embedded and cut, the parts were polished in preparation for FESEM. With regard to the evaluation and the explanations, the spherically shaped green tape structures were virtually divided into different sections, Fig. 4.

The green tape was divided into three main parts, a straight, a crossing and a curved section. The division was necessary because with FESEM only small parts of the cross-section can be analysed in detail. The porosity was characterized by analysing the FESEM images of the different component sections using open source image editing and analysing software (imageJ). The ceramic particles and the porosity had different gray-scale values which were counted. After conversion of the FESEM views into binary images consisting of black ceramic particles and white porosity, the ratio between the number of white pixels and the total number of the pixels was calculated as the porosity.

III. Results and Discussion

Fig. 5 shows a view of a grid-patterned green tape fixed between the hold-down clamp and the matrix (diameter of 62 mm) during deep drawing. In this Fig. 5, the deformation of the green tape can be seen and the grid pattern shows increasing distortion. After drawing, the shaped green tape and the grid visualized the deformation areas and regions of interest.

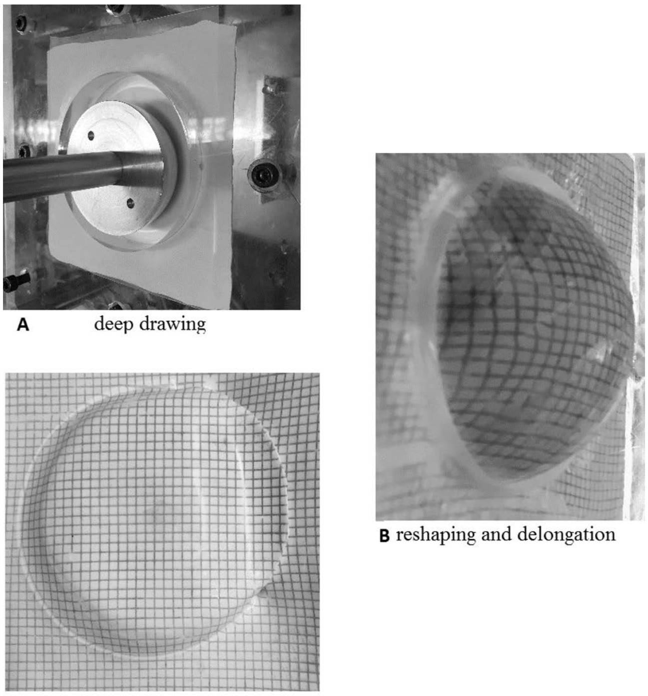

Fig. 5: Deep drawing using a spherical stamp with a diameter of 60 mm (A), visualization of deformation (B) and cracking (C).

The first result of an earlier investigation regarding the influence of drawing speed and relaxation was given by Scheithauer *et al.* [12]. The result of the complete investigation on the deep drawing of ceramic green tape with different parameters, like the solid content, stamp diameter, drawing depth and tape thickness, is summarized in a bar chart, Fig. 6. A correlation between green tape properties and deep drawing results was given for the first time.

The solid content of the green tape influences the properties and therefore it has a significant influence on the forming behavior. Because of the increasing solid content, the resistance against forming increases. The normal binder-polymer has a relative low strength, but with an increase in the solid content, the polymer is supported and therefore the tensile force increases. One the one hand, the ceramic powder particles interfere with the de-looping of the polymer chains. On the other hand, friction increases since more powder surfaces are in contact with each other. In contrast, elasticity decreases with increasing solid content. With decreasing solid content, an increase in the possible drawn depth can be seen as well as with an increasing stamp diameter. The bigger the green tape is, the more material is available for forming the higher drawn depth.

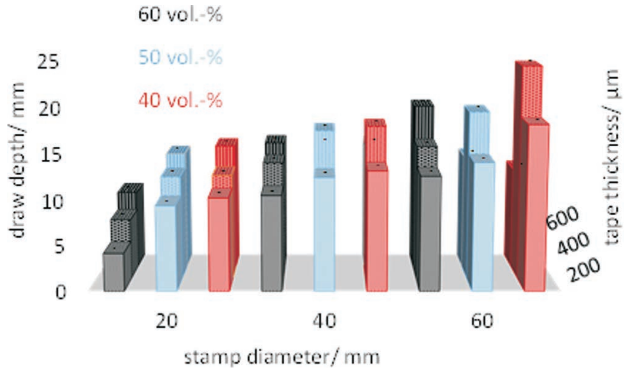

Fig. 6: Summary of the deep drawing result based on comparison of the tape properties and forming parameters.

After deep drawing, the green tape shows deformation easily visualized by the grid pattern. This deformation was digitalised by using a 3D-scan technology (Vialux Company, Chemnitz, Germany), as shown in Fig. 7. With special image processing software, this digital image was evaluated for the calculation of the changes in the mesh of the grid pattern, also visible in Fig. 7.

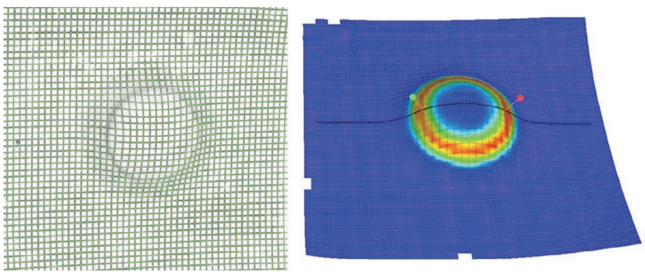

Fig. 7: Digitalized image of 3D-scanned (Vialux GmbH) green tape after deep drawing and marked with a grid pattern.

The digital image shows the deformation of the green tape very clearly and with the software, the elongation in the different forming areas was calculated. Distinctive is a special area in the form of a ring with maximum elongation up to 40 percent. This part of the sample could be critical for thermal processing of the components, but there is no information about the structure and nothing is known about the tape thickness in this area. If the thickness decreases directly with the elongation, homogeneous material shrinkage will occur during sintering. If this is not the case, the particle distance in this area and therefore the local volume powder content have changed, which implies different sintering shrinkage, probably leading to defect and structure tension. The quality of the structure would then be inferior.

In general, sintering of formed tapes was possible for all crack-free drawn tapes independent of the stamp size used. If the drawn depth is 20 mm, the aspect ratio to the stamp diameter increases with decreasing stamp size. However, it is more important to know something about the density within the ceramic structure, especially in the different parts of the formed tape.

Because of this, the described special method for the analysis of different parts of the sample structure was developed. It is important to know the density and particle distribution or distance, especially in a green state and the formed area with maximum elongation, Fig. 7. Fig. 8 shows FESEM views of the different divided parts, non-formed and drawn parts after thermal processing (900 °C) and embedding in epoxy resin for a tape with a solid content of 60 vol%.

The straight area has a structure with a relatively homogeneous particle/porosity distribution. In some areas, individual pores with a bigger diameter can be seen. These pores were caused by air bubbles that could not be completely removed in the evaporation step during slurry preparation. With imageJ, the FESEM views of the structure were analyzed in respect of its porosity. Normally, the porosity of the shown example should be around 40 % because the powder content in the tape was approx. 60 vol%. In Fig. 9, the porosity of all divided areas for described example is given in a bar chart.

In the straight area, the porosity is homogeneous with amounts of 40 % to 45 %, correlating to the theoretical value. In the second "crossing" sector, the result is completely different. For better assessment, the sector is divided into eight sub-areas, each with different porosity. The porosity differs in a range of 40 % to more than 60 % and is very inhomogeneous. Constrictions exist in the outer areas of the crossing section because local elongation limits are exceeded during the drawing (Fig. 8). Sintering of such a structure could be difficult as shrinkage behavior and the total shrinkage differs. Because of the different local shrinkage, tensions can be developed during sintering. In the curve area, the porosity is similar to that of the straight section with values of 43 – 45 %. The results correlate exactly with the 3D-Scan analysis (Fig. 8) and gives an impression of the structural state of the drawn green tape.

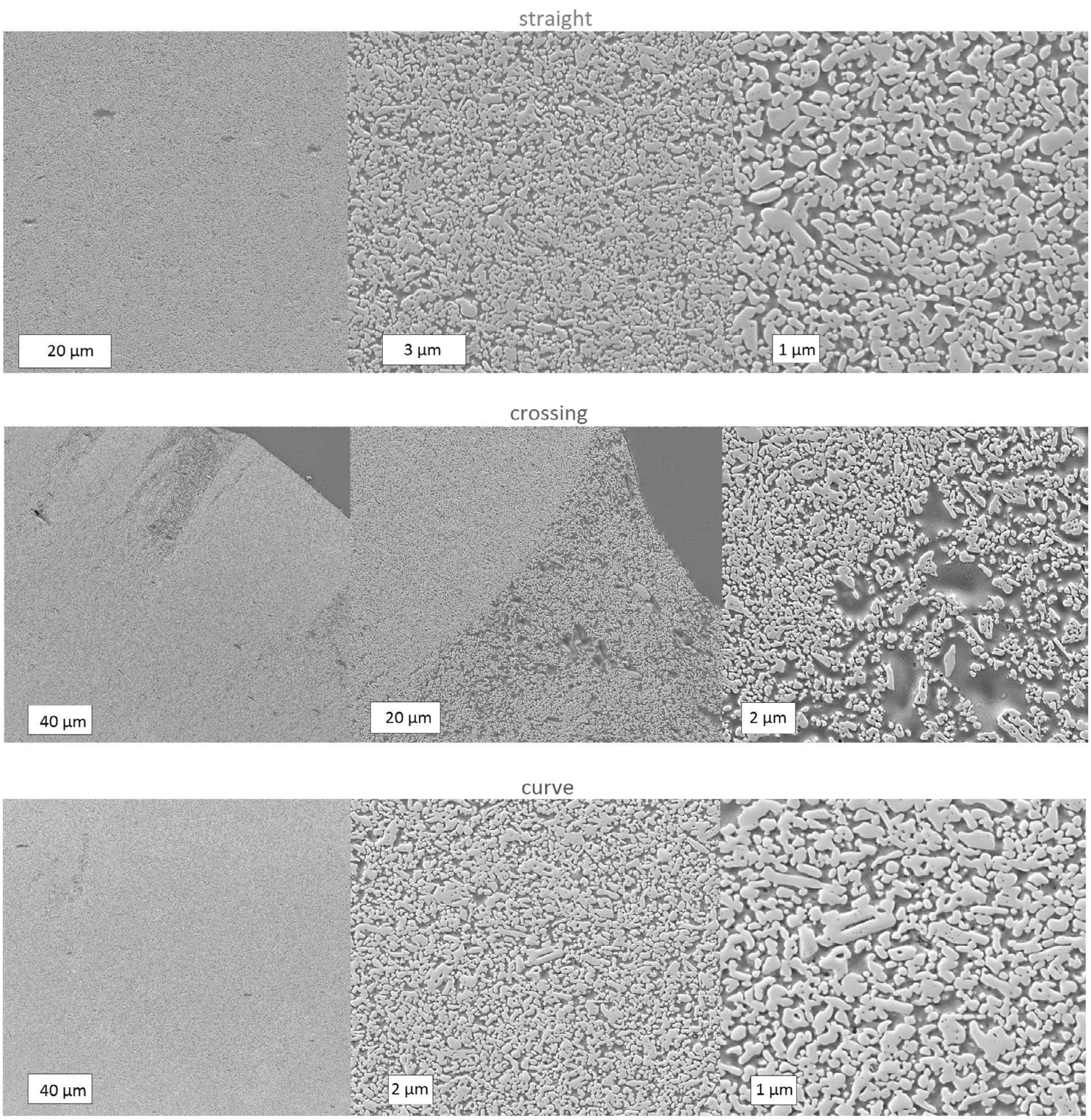

Fig. 8: FESEM of a sintered (900 °C) drawn sample divided in straight (A), crossing (B) and curve area (C).

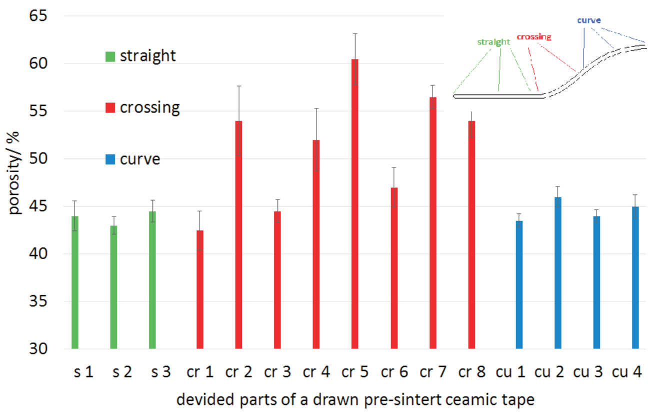

Fig. 9: Summary of calculated porosity in the different areas of the shaped green tape.

With the investigation of the deep drawing for ceramic green tapes, for the first time, new lightweight sintered kiln furniture – a combination of extrudates and green tapes with a nonplanar surface – could be successfully manufactured and sintered without defects, Fig. 10.

Fig. 10: New lightweight kiln furniture (sintered) with a non-planar surface based on a combination of extrudates and ceramic green tapes.

In the first stage, the kiln furniture had a non-planar surface with deep-drawn spherical elements. Two ideas formed the basis for this development – safe sintering of special goods by fixing their position and more effective transport of debinding products thanks to the higher support position. The drawn elements obtained after sintering were completely without defects.

IV. Conclusions

In the metals and plastics industries, deep drawing is well established for the processing of semi-finished planar products to more complex components used in a wide range of application. If deep drawing could be successfully adapted for technical ceramics, many new applications would be possible with the introduction of this technology, for example chemical-resistant housings or new lightweight non-planar kiln furniture. Products of higher complexity, based on ecologically produced water-based ceramic green tapes, could be shaped with the deep-drawing method. For the process, the green tapes need to be prepared in size before deep drawing. Parts of the green tapes may not be usable. From an economic point of view, recycling of expensive materials is a big topic, but, because of the complex recycling process, only ecologically useful for cheap materials. Nevertheless, basic investigations are planned for the future. With this investigation, a basic understanding of the deep drawing of ceramic green tapes could be reached and an in-depth investigation into the mechanism of deep drawing was performed. For the first time, the results show a basic correlation of possible draw depth with the tape properties and deep-drawing parameters. Firstly, a method is presented to investigate the structure of a shaped ceramic green tape. With this method, a more in-depth view into the green structure and the state of ceramic particles, especially the particle distribution, could be gained. Thereby, changes in the structure and porosity in different sections of shaped tapes as critical areas were visualized. The influence and the effect on sintering is another interesting aspect and will be investigated in further studies. Sintering of deep-drawn green tapes was possible, but their structure is currently unknown. The next steps are to examine the lamination of deep-drawn tapes to realize components with a higher thickness. Based on the deep drawing investigation, for the first time, shaped tapes were combined with extruded and machined parts to form non-planar lightweight kiln furniture.

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