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Effect of carbon contamination on the sintering of alumina ceramics

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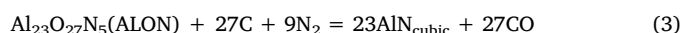
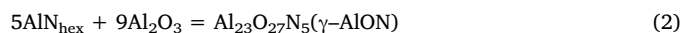
ABSTRACT

The present work aimed with the carbon contamination in alumina ceramics and its influence on sinterability of alumina in low vacuum and atmospheres of argon and nitrogen. The commercially available alumina was coated with carbon and sintered at different atmospheres to investigate the effect of carbon presence on alumina sintering behaviour. The sintering conditions were: heating/cooling rates 5 °C/min and 1.7 °C/min until the maximum temperature of 1400 °C and a dwell time of 2 h. The microstructure of the samples was investigated from fracture and surface, prior to polishing, chemical or thermal etching. The non-densified (porous) surface layer was found in the samples sintered in nitrogen and vacuum, however, sintering in argon atmosphere showed a negligible effect on the surface. The core of investigated specimens exposes a transgranular/intergranular fracture mode and is dense in all cases. In the case of vacuum sintering, the strong carbon diffusivity was also noticeable by the dark grey color of the samples. Interestingly, the formation of aluminium nitride took place during sintering of carbon coated alumina samples in a nitrogen atmosphere at 1400 °C. The thickness of the reactive porous layer was approximately 15 μm beneath the surface. Such a porous layer is inappropriate to the desired features of final ceramic products. Presented results lead to better understanding of the sintering behaviour of ceramic and to suitable selecting of the set-up by densification conditions.

1. Introduction

Alumina ceramics are nowadays densified by various sintering techniques like flash sintering (FS) [1], spark plasma sintering (SPS) [2,3], rapid sintering (RS) [4], pressure-less sintering [5], hot isostatic pressing (HIP) [6], microwave sintering [7] or their combinations [8,9]. Most attention is paid to avoid grain growth and to suppress porosity to a minimum in the dense body. Ceramics prepared in that way can exhibit translucency up to 76.2% at a wavelength of 632 nm, which is used in many fields' e.g. transparent armour or electromagnetic windows [10]. However; ceramic, where optical transparency or translucency is required, is particularly sensitive to carbon contamination which can be introduced during any ceramic processing step (residual organics during powder synthesis due to precursors and ceramic binders, carbon based mixing media, CO – containing atmosphere during calcination and sintering, sintering sleeves and furnace components from graphite, and even mechanical treatment such as machining and polishing) [11]. Moreover, free carbon is not desirable in most of the ceramic materials, as its presence impairs the mechanical properties [12]. Therefore, after the main densification process, the alumina has to be additionally thermally or mechanically post-treated, which is costly, in order to get rid of the carbon contamination or for obtaining a final product with optical quality. But, there is also another issue to be

considered: how the presence of carbon influences the sintering driving forces in the environment with reduced oxygen (oxidation of carbon to CO or CO₂ is suppressed, and carbon is present also during neck formation). M. C. Guenette et al. [13] found that diffusion of carbon (from Ti₂AlC thin films) into a single crystal of α-alumina occurs at temperatures of 570 °C and above. In the presence of nitrogen, aluminium oxynitride or aluminium nitride can be formed by following carbothermal reduction [14]:



As mentioned earlier, alumina is sintered in all type of furnaces consisting of materials which in many cases contain carbon. Often, the final alumina ceramic is planned to be used directly after sintering, without any additional costly mechanical or post-heat treatment procedure. Hence; it is worth to know the possible effect of carbon diffusion on alumina during sintering.

Present work deals with the sintering of alumina in the presence of carbon under vacuum, nitrogen and argon atmosphere. The carbon contamination was simulated via carbon deposition on the pre-sintered samples. For better understanding of the mechanisms and possible

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reactions during densification of the ceramics the samples were also compared without the artificial carbon deposition in case of argon and vacuum treatment. Acquired information could lead to final products without additional mechanical or post – heat treatment steps.

2. Experimental

2.1. Materials and instrumentation

In the present work, high purity alpha alumina powder (99.99%) with mean particle size of ~ 200 nm was compacted in the shape of disc via injection moulding procedure and pre-sintered at 900 °C with a dwell time of 2 h in order to get rid of the carbon in the whole volume of the sample. Such prepared disks with a diameter of 5 mm and thickness of 0.5 mm were carbon deposited. The graphit 33 spray from Buttschardt Electronic AG (Zeiningen, Switzerland) was used. Samples have been sintered at the temperature set-up with a heating/cooling rate of 5 °C/min or 1.7 °C/min up to 1400 °C with 2 h dwell time in atmospheres of argon and nitrogen (60 ml/min) and in a vacuum (10^{-2} mbar), respectively. A tube furnace CTF 17/300 from Carbolite Gero Limited (Hope Valley, United Kingdom) was used for sintering in Ar and N_2 atmosphere. The vacuum sintering was done in a KCE vacuum furnace Sondermaschinen GmbH (Rödentel, Germany) with 10^{-2} mbar vacuum using the same sintering set-up as described above.

The density of the sample was measured on 10 samples via the Archimedes method in distilled water with a measurement uncertainty of less than 0.5%. Relative density was calculated with a theoretical density of utilized alumina of 3.98 g cm $^{-3}$. All measured samples reached $\geq 99.2\%$ of the theoretical density with the deviation among them less than the error factor. Therefore, the relative densities are not further considered or mentioned.

Microstructure analysis was performed with the scanning electron microscopy (VEGA Plus 5136 MM, Tescan instruments, Czech Republic) from the fracture surface as well as from the sample surface on as sintered specimens. The mean size of alumina grains in the sintered samples was determined by the linear intercept method, measuring at least 200 intercepts (software LINCE, TU Darmstadt, Germany), and using the correction factor 1.56 according to [15].

Phase analysis was performed with Panalytical MRX 4 with accelerator detector from PANalytical B.V. (Almelo, Netherland). The source of X-ray radiation was a copper cathode. The diffraction patterns were analyzed by the use of the X-Pert HighScore software.

3. Results and discussion

3.1. Microstructure

The summary of diverse treated alumina ceramics are summarized in Table 1. In order to observe the influence of carbon on sintering behaviour of alumina under different atmospheres, the surface of samples was not mechanically, chemically or thermally treated after sintering.

Samples treated at different atmospheres are shown in Fig. 1

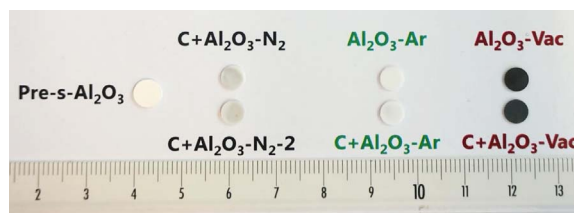


Fig. 1. Pre-sintered sample 900 °C/2 h (Pre-s- Al_2O_3) together with the different thermally treated samples.

together with the pre-sintered sample at 900 °C for 2 h. The deposited carbon on the pre-sintered samples was after sintering removed by ultra-sonication and cleaned in ethanol before analysis. The relative density of all samples thermally treated at 1400 °C for 2 h reached $\geq 99.2\%$ and the differences among diverse treated samples are within the error bar of the measured samples density.

Initially, sintering environment of pure alumina and alumina coated with carbon in vacuum (10^{-2} mbar) is described. It has to be mentioned, that the vacuum furnace has carbon lining and both trials were actually done in the presence of carbon. While in the first case, the carbon is not in direct connect with alumina, whereas the second one has direct contact with carbon. Therefore in Fig. 1 both vacuum treated samples had dark grey colour. The diffusion of carbon into the alumina is related to the decreasing partial pressure of air $p_{(air)}$ during vacuum sintering. Carbon diffusion under vacuum was found in the work of Guenette et al. [13], where the carbon diffusion in a single crystal of α - Al_2O_3 substrate occurs from furnace lining. The diffusion was estimated to be in the order of 3×10^{-3} cm 2 /s in the temperature range of 570 – 770 °C. In the work of Grasso et al. [3], transparent alumina was prepared in a graphite die utilizing the SPS technique at temperatures 950 and 1000 °C under a pressure of 200 – 500 MPa. Resulted samples had a brownish colour which indicates the carbon presence. After annealing for 30 min at 900 °C in air, the brownish colour was reduced and such treated samples obtained an increase in transmittance at 645 nm [3].

The microstructures of vacuum treated samples are shown in Fig. 2. For better comparison, Fig. 2 is divided into three rows with a depiction of the top surface morphology, fracture cross-section close to the surface and fracture cross-section from the bulk of the sample. It can be seen that the microstructure of the sample without carbon layer on the surface had grains with size of 1821 ± 137 nm which is comparable with those found in the core of the sample 1818 ± 172 nm. The fracture morphology exhibits a mixture of trans- and inter-granular fracture. The largest grain size in comparison to samples sintered under nitrogen or argon can be attributed to the overall strong contamination of the utilized furnace previously used for densification of SiC and other materials. Therefore, the overall contamination, with elements supporting grain growth, like silicon, plays an important role. Additionally, bigger grains obtained under the sintering of vacuum were previously attributed to the reduction of partial pressure $p_{(air)}$ during low vacuum sintering as reported in the work of Srdic et al. [16] and Chinellato and Tomasi [17]. However, for grain growth, the impurities present in the

Table 1

Summary of conditions and grain size of the treated alumina samples.

Sample	Heating/cooling rate [°C/min]	Max.temp./dwell time	Carbon	Grain size Surface [nm]	Grain size Fracture [nm]	Colour
Al_2O_3 -Vac	5	1400 °C/ 2 h	–	1821 ± 137	1818 ± 172	Grey
C + Al_2O_3 -Vac	1.7		Spray	NA	2190 ± 141	Dark grey
Al_2O_3 -Ar	5		–	580 ± 71	973 ± 111	White
C + Al_2O_3 -Ar	5		Spray	NA	1020 ± 93	White
C + Al_2O_3 - N_2	5		Spray	NA	719 ± 73	Pale grey
C + Al_2O_3 - N_2 -2	1.7		Spray	NA	1380 ± 60	Pale grey

NA – not available.

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