POROUS Si_3N_4-BASED SUPPORT MATERIALS WITH TAILORED GAS PERMEABILITY

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INTRODUCTION

Porous silicon nitride ceramics can be used for gas separation, also at high temperatures. Silicon nitride has the advantage of chemical stability up to high temperatures and good mechanical properties.[1] For its use as support material in gas separation, gas permeabilities $>10^{-14}$ m^2 are necessary.

The objective of this work was an investigation of the influence of various processing parameters on the permeability behaviour of porous silicon nitride tubes generated via slip-casting.

EXPERIMENTAL PROCEDURES

For all experiments the samples were manufactured via slip casting. The slip contained Si_3N_4, powder (UBE Industries), sintering aids, a dispersing agent and water. As sintering aids Al_2O_3 (Almatis) and Y_2O_3 (H.C. Starck) were used, and Dolapix A88 (Zschimmer & Schwarz) was used as dispersion aid. After casting into plaster moulds, the green bodies were dried overnight at 105 °C followed by sintering at 1600 °C in N_2 atmosphere. Three different Si_3N_4 sources were used; each of them had a different α-Si_3N_4 to β-Si_3N_4 ratio and particle size (Table 1). Mercury porosimetry (Pascal P140 and P440, Porotec), He-pycnometry, water immersion testing, and an air flow test bench were used to investigate porosity, skeleton density, bulk density and air permeability.

Table 1: Particle size and α/β contents of the used powders

	$d_{0.5}$ /µm	α - Si_3N_4 /%	β - Si_3N_4 /%
Si_3N_4-A	1.1	99.4	0.6
Si_3N_4-B	1.6	99.9	0.1
Si_3N_4-C	1.4	88.3	11.7

RESULTS AND DISCUSSION

As a first step, the impact of different concentrations of sintering aids was investigated. As Si_3N_4 source Si_3N_4-A was used. The mass fractions of the sintering aids were reduced subsequently. Starting with the highest amounts of 2.5 % of Al_2O_3 and 2.5 % of Y_2O_3 with respect to silicon nitride, three samples were prepared and sintered at 1600°C. The same was done with mass fractions of 1.0%, 0.5% and 0.2 % of each sintering aid.

Decreasing the amount of sintering aids yielded a higher permeability but reduced mechanical stability. To ensure sufficient stability of the samples, mass fractions of 2.5% of the sintering aids were chosen for subsequent investigations.

To evaluate the impact of Si_3N_4 source, samples made of Si_3N_4-B and Si_3N_4-C powders were sintered at 1500 °C, 1550 °C and 1600 °C. A difference in the morphology of the microstructure depending on the powder type was observed using scanning electron microscopy. According to the literature [2], this is caused by the different amounts of β-Si_3N_4 in the starting materials.

A lower sintering temperature leads to a higher permeability. For Si_3N_4-C sintered at 1500 °C and 1550 °C, a high scattering of permeability values was observed. The samples made of Si_3N_4-C and sintered at 1600 °C yielded a higher permeability than the samples made of Si_3N_4-B regardless of the sintering temperature.

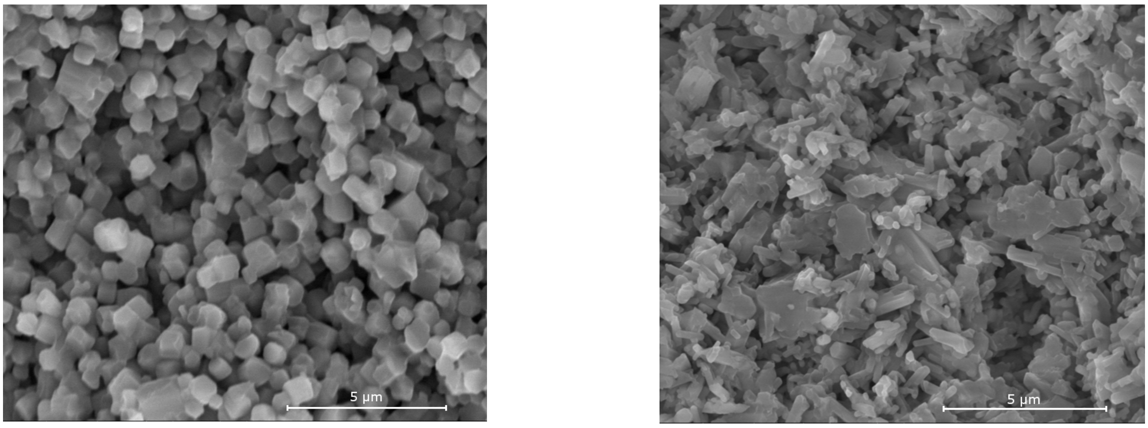

Fig. 1: left: fracture surface of Si_3N_4-B with equiaxed grains, 0.1 % β-Si_3N_4 in the starting powder; right: fracture surface of Si_3N_4-C with anisotropic rod-like grains, 11.7 % β-Si_3N_4 in the starting powder

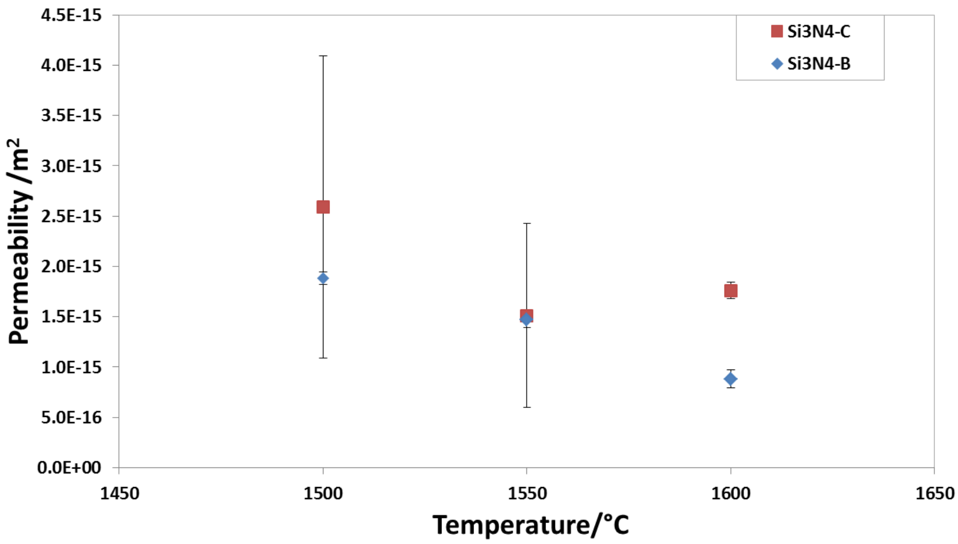

Fig. 2: Permeability as a function of sintering temperature for Si_3N_4-B and Si_3N_4-C. Each point is the mean value of four samples.

CONCLUSIONS

The impact of different processing parameters on porosity and permeability was shown. Based on the data at hand, a sintering temperature of 1600 °C with 2.5 % of both Al_2O_3 and Y_2O_3 as sintering aids with Si_3N_4-C as Si_3N_4 source was most promising. At the moment the desired permeability of 10^{-14} m^2 could not yet be achieved, thus requiring further work.

REFERENCES

[1] Konegger, T., et al. (2016). "Asymmetric polysilazane-derived ceramic structures with multiscalar porosity for membrane applications." Microporous and Mesoporous Materials **232**: 196-204.

[2] Kalemtas, A., et al. (2013). "Mechanical characterization of highly porous β-Si3N4 ceramics fabricated via partial sintering & starch addition." Journal of the European Ceramic Society 33(9): 1507-1515.