COATING OF TUBULAR SILICON NITRIDE SUPPORT STRUCTURES WITH POLYMER DERIVED CERAMIC MEMBRANE LAYERS

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INTRODUCTION

Non-oxide ceramic membranes boast chemical and thermal stability which makes them suitable for the use in many of today's membrane processes. Because of highly beneficial mechanical and thermochemical properties, porous silicon nitride[1],[2] in combination with polymer derived SiCN [3] can be used as membrane materials, suitable for prospective uses under harsh circumstances that common polymer- or metal-based membranes cannot withstand.

The objective of this work is the development of a processing routine for preparing asymmetric membrane structures, using tubular macroporous silicon nitride supports in combination with polymer derived Si(CN) membrane layers deposited via a dip coating process similar to what Konegger et al. have described [4]. The final layer needs to be microporous (pore diameter smaller than 2 nm). Porosity can be achieved by different processes. For the macroporous support a partial sintering route was chosen, the viability of which has been demonstrated previously [5], while the microporous separation layer is produced by pyrolysis of a preceramic polymer.

EXPERIMENTAL PROCEDURE

In this work, two different coating approaches were evaluated. The first approach included an application of a mesoporous intermediate layer onto the substrate before coating it with the separation layer, while the second approach involved the direct deposition of the separation layer onto the supports.

Preparation of support structures: The tubular macroporous support structures were infiltrated with polystyrene solution (25 wt% PS in toluene) by a dip-coating process. Subsequently, the surface of the masked support tubes was polished in order to remove coating flaws.

Deposition of the intermediate layer: An intermediate layer was prepared by dip-coating the masked support structures in dispersions consisting of an organic solvent (primarily n-hexane, which does not dissolve the masking compound and does not react with the preceramic polymer), silicon nitride (SN-E10, UBE) and poly(vinyl)silazane (Durazane 1800, durXtreme GmbH). Here, the poly(vinyl)silazane compound serves as a binder. After dip-coating, the samples were crosslinked in a furnace at 130 °C under flowing nitrogen. The masking compound was dissolved in toluene before the samples were pyrolysed at 600 °C under flowing nitrogen.

Deposition of the top layer: The separation layer was either coated onto the intermediate layer or directly coated onto the masked supports by the same dip-coating procedure as before, using a solution of the preceramic polymer in an organic solvent. After crosslinking (130 °C, nitrogen) the masking compound was dissolved in toluene and the sample was pyrolysed (600 °C, nitrogen).

RESULTS AND DISCUSSION

As no stable dispersions of polymer, silicon nitride powder and compatible organic solvents could be prepared, the dispersion had to be continually stirred, which makes predicting coating behavior challenging, due to it not being a stable system. In spite of these challenges, continuous intermediate layers were successfully deposited onto the masked support structures.

An almost defect free top layer was achieved by direct coating of the masked supports (Figure 1). The number of deposition defects can be reduced by multiple coating runs. However, many variables have to be considered during the whole production process, which can influence the quality of the top layer.

Coating onto the intermediate layer appears more promising, since the surface of the layer is smoother and more homogenous than that of the support. However, development of the coating process has to be continued in order to obtain surface qualities suitable for membrane applications.

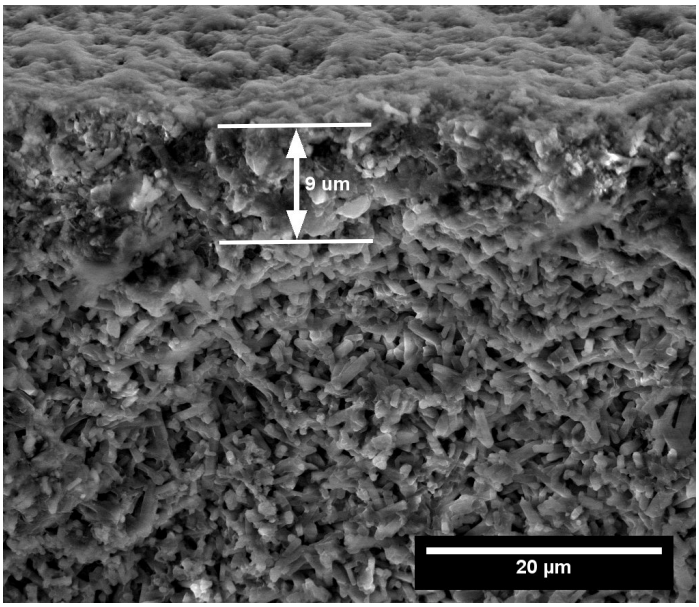

Figure 1: SEM micrograph of top layer surface after a singular coating step directly onto the masked substrate surface.

CONCLUSIONS

The coating of macroporous silicon nitride support structures with a polymer derived SiCN membrane layer of a few µm thickness was achieved using a dip coating process. An optimized masking technique allowed for a direct coating of the supports, yielding an almost crack-free top layer. An improvement of the surface quality of the substrate was achieved by adding an additional intermediate layer to the process. The results are promising and therefore, the focus lies on further continuing the deposition process development.

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