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Mechanical testing of adherence of stacked layers in tubular geometry

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Abstract

For the development of new molecular separation technologies strong robust tubular membrane systems are required. The fragile membranes, however, need a strong defect free support such as a porous asymmetric ceramic tube. Mechanical failure of these ceramic membrane systems during manufacturing and operation is mainly caused by delamination of the stacked layers. Therefore development is focused on improving the adherence. As no standard mechanical test for tubular samples is available yet, a new tensile test was developed to facilitate the current research. The most important components in the new equipment is a test tool with a curvature matching that of the test sample and a sample casing that align and guide the test tool during the tensile test. With this tensile test the manufacturing procedure for the ECN standard tubular α-alumina support was optimized. Firing the asymmetric support at 1300°C resulted in the highest mechanical strength for the support system with cohesive fracture in the support tube. With the test developed the process condition could be identified where the material of the support tube is the weakest link in the support system.

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1. Introduction

At ECN in the unit Energy Efficiency in the Industry (EEI) one of the research lines is the development of tubular microporous ceramic membranes for several molecular separations such as pervaporation, gas separation and nanofiltration. The pore size of the functional membrane is only a few Ångstrom, and to reach a high gas flow through the robust membrane system, very thin layers of only a few hundred nanometers are required. However, these layers are very fragile and are therefore deposited on a support that needs to be strong with an appropriate pore size and low flow resistance. The support system under development is an asymmetrical support consisting of a strong macroporous support with several thin layers of different pore size [3, 4]. The adhesion between these layers and the support is one of the key issues in manufacturing reliable support systems. Adhesion is realised by sintering, densifying, the ceramic support at elevated temperature. However, densification of these layered systems is retarded by constrained sintering [1] and high temperatures are needed. This results in increasing shrinkage stresses, tensile stress, and consequently higher probability of cracking and delamination of the layers.

For certain industrial applications, tubular membranes were shown to be the most promising geometry [2]. During operation the membrane and support system is subjected to both internal or external pressure and residual stresses from the fabrication process. Hence, for a reliable operation of tubular membranes, the cohesive and adhesive strengths of the asymmetric support system are of great importance. To determine whether the support system can withstand these stresses - tensile, compressive and shear - the cohesive and adhesive strength should be characterized. During operation of the tubular asymmetric membrane the primary load mode that may result in spallation of the layers is the internal pressure. Tensile testing could simulate such an internal pressure and enables studying the effect of system residual stresses (sintering) on the cohesive and adhesive strength of the layered structure. The standard tensile test [5], although relatively easy to perform using standard mechanical testing equipment, is restricted to flat geometry.

The goal of the present study is the development of a tensile test for tubular geometry enabling the determination of the optimal sinter temperature for manufacturing strong reliable asymmetric supports for molecular separation processes. Here, we will not discuss the influence of the sinter temperature on other important parameters such as pore size distribution, defect concentration etc of the resulting layer.

2. Experimental procedure

The tensile test that is developed for testing the adherence of stacked layers is based on the ASTM Standard test method for Tension testing [6]. The new equipment had to meet the following objectives:

- the curvature of the test tool has to match that of the tubular support
- the porous sample has to be fixed to the test tool without severe penetration of the glue into the porous material
- the sample glued onto the test tool has to be fixed in the testing equipment without premature delamination
- the alignment of the test tool must assure solely tensile stresses.

Several concepts have been evaluated for the test tool taking into account: the brittleness of the ceramic sample, unroundness of the tube and surface roughness. The final design was a concave curved test tool with a small circular cross section and a sample holder that restricts the movement of the test tool to only the y-(tension) direction.

Asymmetric supports are manufactured by colloidal processing according the processing flow sheet in figure 2.1.The porous support tube is a commercial α-alumina tube, inner/outer diameter of 10/14 mm. Thin porous layers of 45 µm each are applied by a film coat technique developed at ECN, using α-alumina suspensions of particles of 600 nm. After film coating the support system is dried and sintered in air for 2 hours. A more detailed description of the preparation method can be found in [3]. The effect of the sinter temperature on the adherence was studied in the range of 1100°C to 1300°C.

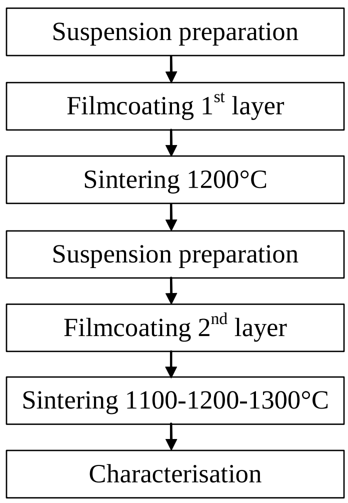


Figure 2.1 *Processing flow sheet for standard ECN support system*

3. Results and Discussion

3.1 Test equipment and procedure

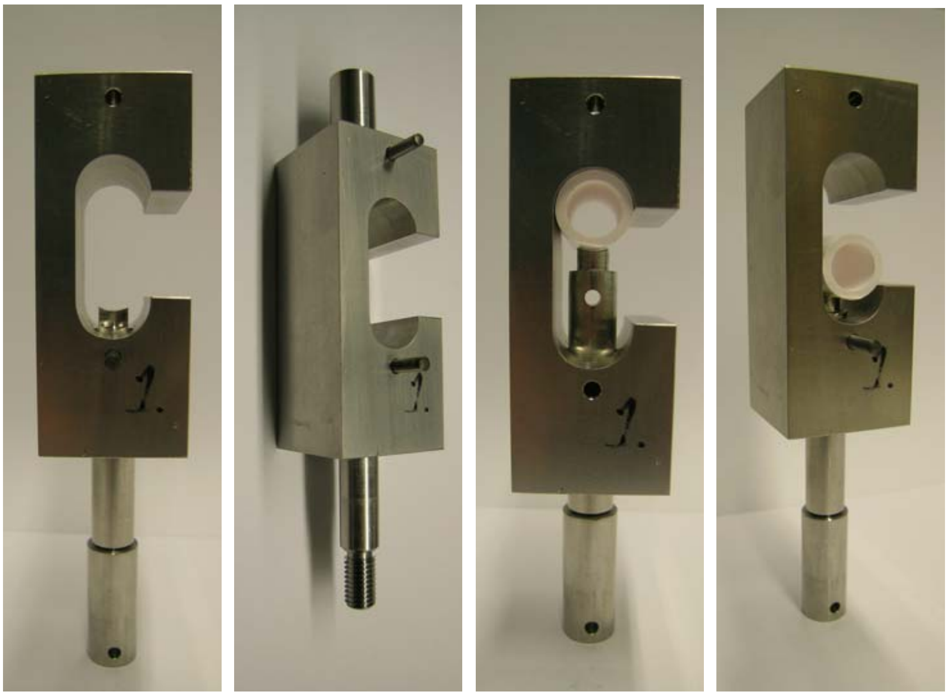
The final tensile test equipment is illustrated in Figure 3.1 *Test tool and holder for tensile testing of tubular sample* and consists of two basic components that is:

- the test pin (tool) with a curvature matching that of the sample tube and
- the sample casing (holder) with two functions, first fixation of the test sample and test pin for hardening of the glue and second restricting the movement of the test pin in the tensile mode of testing.

To attach the test sample to the tool and execute the tensile test a special holder was designed. In designing the holder special attention was paid to the alignment of the sample and guidance of the tool during the tensile test, see Figure 3.1.a . The test samples are mounted on the tool using a cyanoacrylate adhesive. Preliminary testing showed that there is only a restricted penetration of the adhesive into the porous sample, minimizing the influence of the adhesive on the test.

The test procedure is as follows and visualized in Figure 3.1. The surfaces of the tool and test sample are cleaned with clean pressurized air to remove dust. Then the tool and the holder are assembled, Figure 3.1.a , and the position of the tool is fixed using a fixation pin, see Figure 3.1.b. The test sample is placed in the upper position for mounting and the fixation pin for the tool is removed. The cyanoacrylate adhesive is applied on the tool surface and evenly distributed over the surface. Immediately after applying the adhesive, the holder and test sample are lowered and the adhesive is allowed to dry for 30 minutes. After 30 minutes the load is removed and the tool with the test sample glued on it are fixed in the bottom position of the holder, see Figure 3.1.d. After a 24 hour post cure, the holder, tool and sample are placed in the mechanical testing machine, Figure 3.2. After removing the fixation pin the tool and sample are lowered just above the tensile position. Then the tensile test is performed in the strain rate controlled mode (0.5 mm/min) while recording the displacement and load.

Preliminary testing showed perfect brittle fracture for the tubular ceramic supports, indicating a proper alignment of the test tool.



a b c d

Figure 3.1 *Test tool and holder for tensile testing of tubular samples*

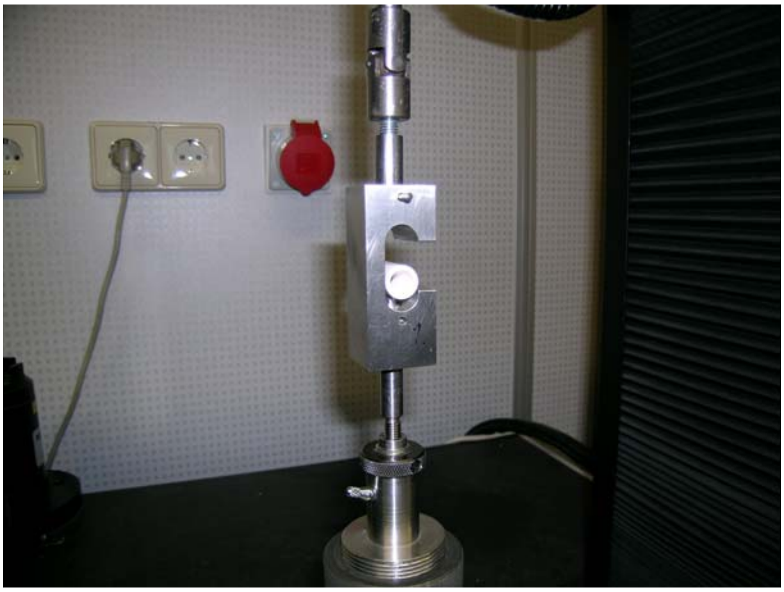


Figure 3.2 *Tensile test set up as developed at ECN*

After tensile testing the fracture surface on both the tool and the sample are analyzed. Characterization is focused on the failure location and failure mode: tool/glue, cohesive in either one of the layers or the support, adhesive via one of the layer interfaces or the interface with the support. If failure of the tool/glue interface occurs and the failure load is below the failure load of the adhesive the test is considered as invalid.

3.2 Optimization adherence in asymmetric support

Figure 3.3 shows the stress-strain diagram as measured during testing of the asymmetric supports sintered at different temperatures. The diagram shows a perfect brittle fracture with a wide variation in strength, see Table 3.1. This behavior is typical for ceramic materials and all the samples tested showed a brittle facture. By increasing the sinter temperature from 1100 to 1300ºC the adherence of the layers in the asymmetric support increases from 10 to maximal 300 N, see Table 3.1.

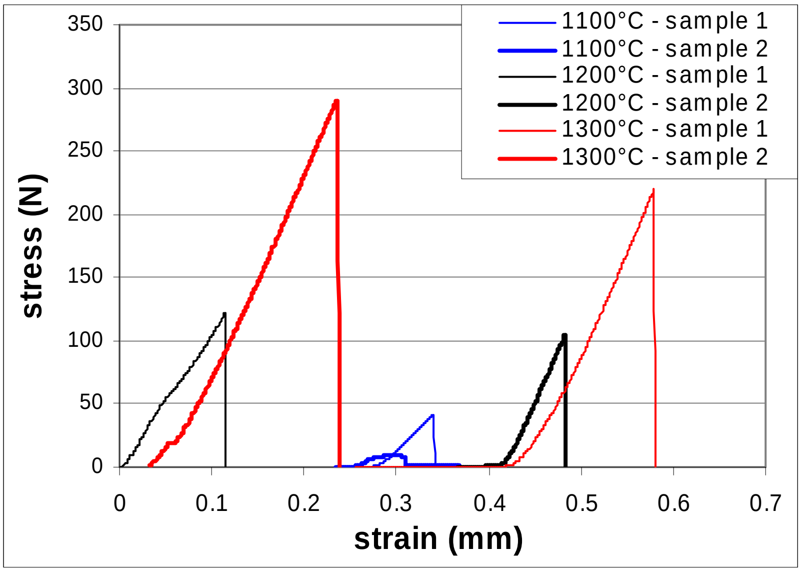
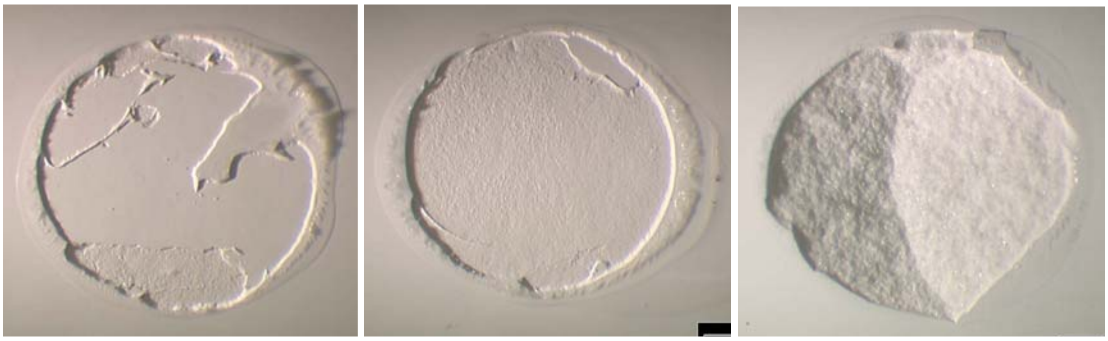


Figure 3.3 *Stress-strain diagram for the asymmetric supports sintered at 1100, 1200 and 1300ºC.*

Fractography reveals the weakest link in the asymmetric support as can be seen in Figure 3.4 and Table 3.1. By increasing the sinter temperature from 1100 to 1300ºC failure of the multilayer structure occurs deeper into support system. Ultimately, at sinter condition 1300°C, the bulk material of the support tube showed to be the weakest link in the asymmetric support.

Table 3.1 *Sinter temperature and adherence of the layers in the sintered support*

Sinter temperature (°C)	Adherence [N]	Weakest link Fracture
1100	10-40	Interface 1st and 2nd layer
1200	104-122	Interface support tube and 1st layer
1300	220-290	In bulk material support tube



a b c

Figure 3.4 *Fractography of the fractures surfaces: a, 1100ºC - interface 1st and 2nd layer, b = 1200ºC – interface support tube and 1st layer, c = 1300ºC – bulk material support tube*

4. Conclusion

The new tensile test equipment and test procedure showed to be a suitable tool for characterizing the adherence of stacked layers in a tubular geometry. At this stage of development of the test only a maximal load is obtained. But still the technique facilitates the optimization of the manufacturing process for strong reliable multilayered support systems by detecting the process condition where the support tube becomes the weakest link. For the standard tubular ECN α-alumina support system the sinter temperature at which the material of the bare tube becomes the weakest link is identified as 1300°C.

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