High Efficiency Recycling and Reusing of Laundry Wastewater by Ceramic-Membrane Nanofiltration

The continuous rise in living standards and the growth of industrial water usage strongly increase the demand for freshwater supply. Consequently, further development of applicable technologies that facilitate internal industrial wastewater treatment is much needed. Over the last decades, some noticeable improvements have been recorded in membrane filtration technologies.

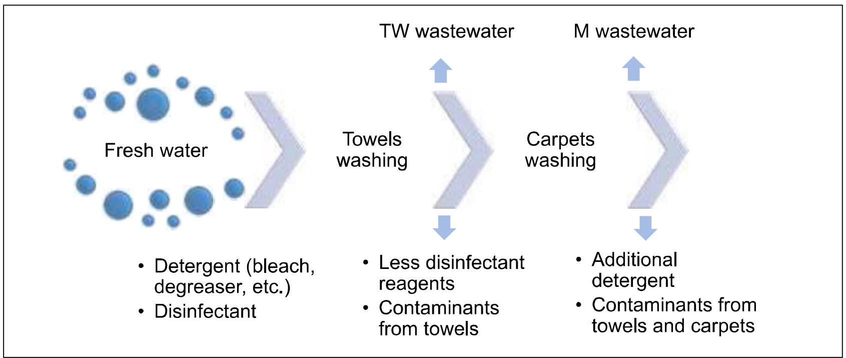

Fig. 1
Schematic representation of the laundry process and wastewater sampling points, TW-towels wastewater, M-carpets wastewater

Introduction

inopor®, as a Rauschert brand and producer of ceramic membranes, offers not only the membranes, but also the service of customised optimization of technology with a key focus on membrane filtration.
The sustainable treatment mechanisms of these technologies, reduced energy requirements and compact equipment sizes benefit the water and wastewater treatment process.
The introduction of wastewater recycling systems into the laundry industry is an important sustainable development, which will positively influence the global water scarcity situation. A couple of treatment technologies can be chosen for this application, but ceramic-membrane filtration, from inopor® for example, is more suitable. Advantages include its compactness, the avoidance of additional chemicals such as flocculants (with the exception of membrane cleaning cycles) in the wastewater treatment and recycling, and the long service life of the systems and membranes. Because inopor®-membranes are ceramic membranes, direct filtration in the hot chemical-water cycle is possible. This brings an additional energy saving effect, because cooling of the wastewater before recycling is not necessary.

Keywords
ceramic-membrane nanofiltration, recycling of laundry wastewater, recycling of laundry cleaning solutions

Current situation and objective

As a first step of the ReWaMem project, which aims to improve the sustainability of the laundry process at CHMS (Coburger Handtuch+Matten-Service), the set of wastewater treatment experiments were performed at the Rauschert Kloster Veilsdorf site.
CHMS specialises in laundry of towels (TW) and carpets (M); they work with customers from different industries and countries. Since there is no strict regularity in the delivery of carpets and towels to CHMS, the quality of the produced wastewater may vary from wash to wash. As a consequence of a significant variation in the pollution sources, the wastewater contained different inorganic and organic matter. Apart from the removed contaminants, added detergent and disinfectants can influence the wastewater quality, e.g. by raising the COD content. The laundry process is performed in a batch mode, where first the freshwater is applied for towel washing and, as a following step, the produced wastewater is used for carpet washing (Fig. 1). Both wastewater samples were collected and treated separately. The volume flow of the wastewater production is around 50 m³/day.

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As the main aim of this project is to be able to treat produced wastewater and recycle it, there are quality requirements that have to be met. The complete information on the water quality recycling limits is summarised in Tab. 1.

Tab. 1.
Water quality requirements for the recycling application at CHMS

	Quality Standard	Recycling limit
In situ parameters	Electric conductivity [µS/cm]	400
	pH	7–10
Sum parameter	COD [mg/l]	–
	Heavy metals:	–
	Pb^{2+} [mg/l]	0,5
	Cu^{2+} [mg/l]	0,5
	Zn^{2+} [mg/l]	2

Preliminary examination

Based on the project objectives, the test plan was created to analyse a broad variety of ceramic membranes and their treatment characteristics with a specific laundry wastewater. The series of tests, with single and multichannel inopor® membranes, were carried out with the application of VSA, a lab-scale filter system, and InoMini laboratory filter systems (Fig. 2 and Fig. 3).

Tab. 2
Tested membrane geometries and their properties

Geometry	Flow Channel Diameter [mm]	Number of Channels	Outside Diameter [mm]	Filtration Area [m²/m]
AA	7	1	10	0,022
CA	3,5	19	25	0,209
HA	2	163	41	1,098
NA	2	151	41	1,072

Laboratory test trials

Variation in membrane geometry and treatment conditions

inopor® offers different ceramic membranes in a wide variety of geometries for industrial applications (Overview at www.inopor.com). For this project, 1-, 19-, 151- and 163-channel membranes were applied for the laundry wastewater treatment (Tab. 2). In addition, for each geometry, two different Al_2O_3 membrane supports (A3/A4) and TiO_2 membrane coatings (T-200/T-450) were examined. For each type, three individual membranes were examined. The aim of this step was to determine the membrane which would achieve the best treatment results and

Fig. 2
VSA lab-scale treatment unit

Fig. 3
InoMini laboratory cross-flow filter system

Tab. 3
Laundry wastewater quality analysis

	Parameter	N	M wastewater			N	TW wastewater		
			Min value	Mean value	Max value		Min value	Mean value	Max value
In situ Parameters	pH	100	6,7	7,8	9,8	100	8,0	9,9	11,5
	EC [µS/cm]	100	1900	2221	2900	100	1900	2206	2600
Sum parameters	COD [mg/l]	100	2100	2742	3100	100	3100	4036	5200
Cations	Na^+ [mg/l]	1	374	374	374	1	488	488	488
	K^+ [mg/l]	1	45	45	45	1	72	72	72
	Cu^{2+} [mg/l]	18	0.20	0,27	0,33	15	0,26	0,34	0,72
	Zn^{2+} [mg/l]	18	2,40	3,40	4,80	15	0,88	1,05	1,30
	Pb^{2+} [mg/l]	18	0,08	0,09	0,13	15	0,05	0,06	0,08

could be further applied to a production plant. To establish the most suitable operational pressure, two different TMP's (trans membrane pressures) were tested with multichannel membranes (10 bar and 20 bar). With respect to the single-channel membranes, a TMP of 30 bar was also examined.

Variation in the laundry wastewater quality

During the treatment process, two different types of laundry wastewater were analysed at the Rauschert laboratories towels (TW) and carpets (M) wastewater. From the water quality analysis, it was determined that the wastewaters exhibit slightly different characteristics. Compared to M wastewater, TW contained significantly higher organic content (COD up to 5200 mg/l); additionally, the pH of this water type was more basic. All the analysed parameters were summarised in Tab. 3.

Laboratory tests

Treatment experiments were performed with the use of both types of wastewater, to analyse the membrane treatment efficiency. Membranes were not chemically cleaned or backwashed between the filtrations.

The tests were carried out at 10, 20, and 30 bar trans-membrane pressures in feed-batch mode, with crossflow velocities of 4,0 m/s and 3,0 m/s. The system was cooled throughout the filtration process to maintain a stable feed temperature of 20 °C.

After performing preliminary trials with the single-channel membranes and analysing the results, it was decided to shift the focus to the membrane geometries of a greater membrane area (multichannel membranes). A higher membrane area allows greater wastewater volumes to be treated per unit of time, which is preferable for the industrial application.

Permeate flux

Fig. 4 illustrates the permeability of the multichannel membranes. Each box plot includes 3 values obtained from three tested membranes of the same membrane pore size and same support material. As can be inferred from the graph, the 151- and 163-channel membranes had the highest deviation between permeability values, while, in the case of 19 C-membranes, the outcomes were mostly more consistent (Fig. 4).

Additionally, a slight variation can be seen between different membrane supports (A3

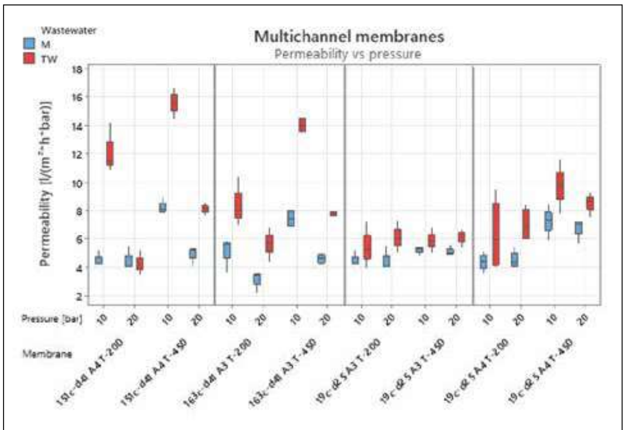

Fig. 4
Permeability outcomes collected from the multichannel membranes at TMP of 10/20 bar, carpets wastewater (M), towels wastewater (TW)

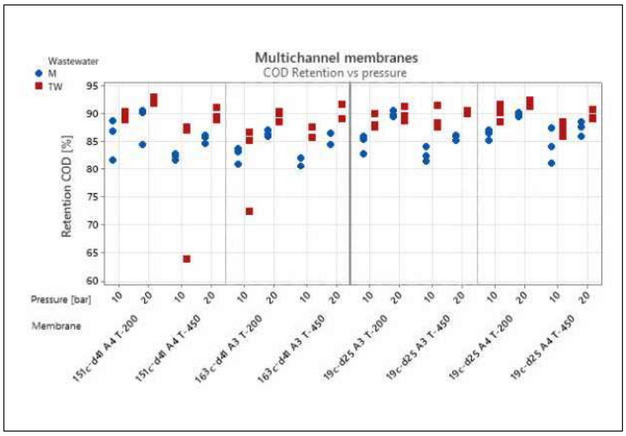

Fig. 5
COD retention of the multichannel membranes at TMP of 10/20 bar, carpets wastewater (M), towels wastewater (TW)

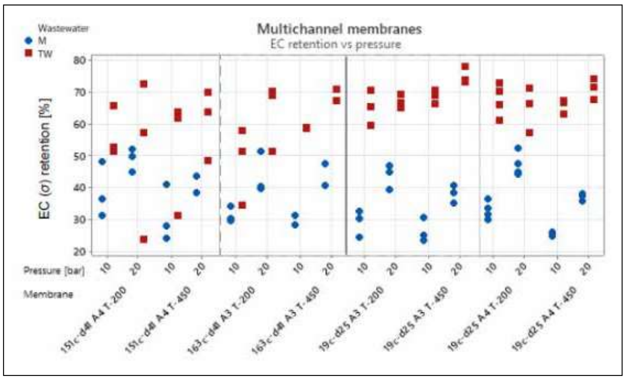

Fig. 6
EC results obtained from multichannel membranes at TMP of 10/20 bar, carpets wastewater (M), towels wastewater (TW)

Tab. 4
Concentration of monovalent ions in the permeate produced by multichannel membranes, carpets wastewater (M), towels wastewater (TW)

Membrane	Parameter	Permeate	
		M wastewater	TW wastewater
151C-d41 A4 T-450	Na^+ [mg/l]	252	145
	K^+ [mg/l]	26	16
151C-d41 A4 T-200	Na^+ [mg/l]	189	100
	K^+ [mg/l]	21	14

vs. A4). A small advantage of the A4 support can be observed. This trend is noted for all the multichannel membranes, highlighting the influence of the support material regardless of the geometry of the membrane. Similarly, the influence of the membrane coating can be seen. Membranes with the T-450 coating reached higher flux, compared to T-200.

COD retention of the multichannel membranes

Fig. 5 represents the COD retention of the tested membranes. Each dot represents a single value collected from an inopor® membrane of the defined type. For each type, three individual membranes were examined.

On average, all the multichannel membranes showed retention in the range of >80 %. A retention of up to 93 % was observed during the tests, however this was heavily dependent on the initial COD content of the laundry wastewater. Regarding this parameter, no significant influence of the membrane geometry (19C-,151C-, 163C-) was recorded. A comparably efficient retention was achieved regardless of the membrane area.

Similarly, the retention efficiency was not influenced by the membrane type (T-200 vs T-450) or support material (A4 vs A3) – T-200 vs T-400 stands for the industrial nanofiltration membranes with the cut-off 200 Da vs 450 Da [https://inopor.com/en/products/membranes.html]. It can therefore be concluded that the organic matter impurities present in laundry wastewater are large enough to be equally removed by all the membrane types.

Electrical conductivity (EC) retention

Another studied parameter – the Removal Efficiency (EC), was recorded at up to 78 %. However, in the majority of the studied cases, the required limit of 400 µS/cm was not achieved.

The EC in the permeate was in the range of 500–1500 µS/cm. It is suggested that the desired limit was not reached due to the inability of NF alone to completely retain monovalent ions.

After a set of water quality analyses, an elevated concentration of monovalent ions was recorded in the permeate. These results highlight that the monovalent ions can be retained with multichannel NF, but not completely (Tab. 4).

Throughout all the treatment tests, a clear difference was recorded between the two types of wastewater. An obvious advantage for TW was observed, it was suggested that the water quality was the reason behind this behaviour, as the process conditions were identical regardless of the water type.

Heavy metals removal

As the main pollutants of concern in the laundry wastewater are heavy metals in terms of Zn^{2+}, Pb^{2+}, and Cu^{2+}, the permeate samples were tested for their presence. After the comparison of the initial and final concentrations, it was determined that the retention is in the range of 97 % to almost 99 % for all the investigated metals (Tab. 5).

Based on the outcomes collected from the single-channel membranes, there is no

Tab. 5
Heavy metal concentration in the permeate produced by single-channel membranes, carpets wastewater (M), towels wastewater (TW)

Membrane	Permeate	TMP [bar]	Pb^{2+} [mg/l]	Cu^{2+} [mg/l]	Zn^{2+} [mg/l]
1C-d10 A3 T-200	M	10	<0,01	0,03	0,03
		20	<0,01	0,02	0,02
		30	<0,01	0,02	0,02
	TW	10	<0,01	0,02	0,02
		20	<0,01	0,01	0,01
		30	<0,01	0,02	0,01
1C-d10 A3 T-450	M	10	<0,01	0,02	0,02
		20	<0,01	0,02	0,02
		30	<0,01	0,01	0,01
	TW	10	<0,01	0,02	0,02
		20	<0,01	0,02	0,02
		30	<0,01	0,01	0,02

Tab. 6 Heavy metal concentration in the permeate produced by multichannel membranes, carpets wastewater (M), towels wastewater (TW)

Membrane	Parameter	Permeate	
		M Wastewater	TW Wastewater
151C-d41 A4 T-200	Cu^{2+} [mg/l]	<0,01	<0,01
	Zn^{2+} [mg/l]	<0,01	0,04
151C-d41 A4 T-450	Cu^{2+} [mg/l]	<0,01	<0,01
	Zn^{2+} [mg/l]	<0,01	0,02

significant influence recorded for either the support layer (A3 vs. A4), or coating (T-200 vs T-450). Outstanding removal efficiency was achieved by all the tested inopor® membranes, reaching the required limits.
The permeate produced by multichannel membranes was also examined for heavy metal content (Tab. 6). Similarly to single-channel membranes, pollutants of concern were retained efficiently.

Summary of the results

All the tested inopor® multichannel geometries, support layers and membrane coatings are potentially suitable for full scale application, showing comparably high results.
The 151C- and 163C- membranes exhibit higher membrane areas, therefore they are preferred for the application. Additionally, the A4 support material and T-450 membrane coating were chosen over A3 and T-200, due to lower investment costs. Thus, based on all the parameters, the optimised final variant is selected to be 151C-A4-T-450.

Optimization

Determination of the optimal operating point

Based on the laboratory test results, the optimum operating pressure was determined to be 20 bar. At this pressure, efficient retention of the target compounds (COD, EC, heavy metals) was achieved, and the membrane flux remained high.

Filtration tests under production conditions.

As part of the investigation, several experimental tests were performed to gain a more extensive picture of the membrane filtration behaviour and to elaborate on its full potential. The filtration conditions were modified, with both the treatment temperature and filtration time increased.

6.1 Treatment temperature effect on the permeability

The test with increasing temperature, performed with a single-channel inopor® membrane, was aimed to analyse the correlation between the increase of the feed temperature and the permeability (Fig. 7). The results revealed that as the temperature was raised, the permeability increased. During the filtration, water was heated from 20 °C to 55 °C, and the permeability increased from 6,1 l/(m² · h · bar) to 13,9 l/(m² · h · bar). This happened due to the decrease in the viscosity of the feed wastewater, and that led to a water flux increase. These results show that in the full scale application, where high temperatures of ~ 40–50 °C are applied for washing, this membrane behaviour will benefit the operation process.

Repetitive filtering

This test was performed with the 163C-d41 A3 T-450 inopor® membrane with the use of the VSA equipment. The filtration process took 7 h at a TMP of 20 bar, during which the wastewater was changed six times. The treatment temperature ranged between 30–45 °C due to a lack of cooling system capacity. The presented permeability values were estimated excluding temperature. However, the elevated temperature was beneficial, as in the real-case application, higher temperatures are applied (over 45 °C) due to the washing temperature needed for the laundry process. No membrane cleaning was performed in between the feed changes; therefore it was possible to estimate the fouling rate of the membrane.
Based on this, it was confirmed that the water quality has an influence on the membrane performance. In the case of the TW feed, the flux was higher compared to M. However, regarding the

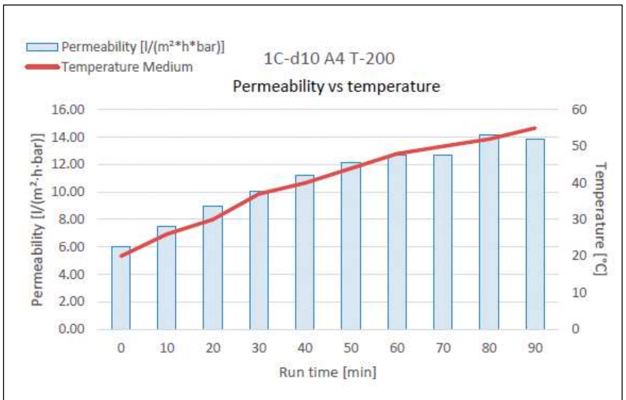

Fig. 7
Temperature and membrane permeability correlation with a single-channel membrane

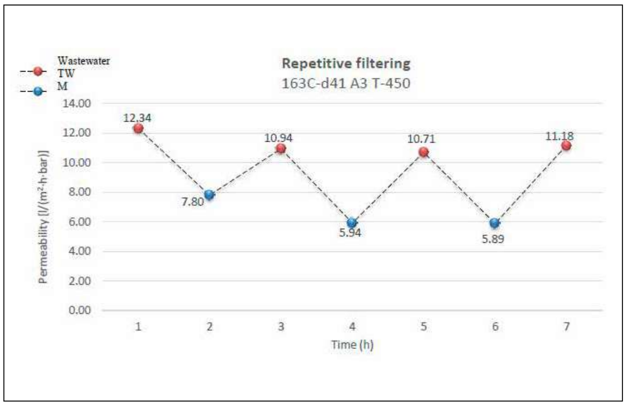

Fig. 8
Repetitive filtering results for 163C-membrane, at TMP of 20 bar, carpets wastewater (M), towels wastewater (TW)

membrane permeability, very satisfactory results were achieved, showing stable performance throughout the 7 h filtration process.

Solutions for the further application
In relation to the quality limits required for the recycling application, the EC values in the permeate are not reached with the application of single NF membranes. The solution, which can be utilised in order to lower the EC in the produced permeate, is a simple dilution with fresh water. The addition of fresh water will be required for the industrial application anyway, as the amount of produced permeate will be lower than the required water volume for the laundry. Therefore, the limit of 400 µS/cm can be reached via dilution.

Summary and next steps
A broad variety of laboratory experiments provided a substantial overview of the behaviour of the ceramic membranes during laundry wastewater treatment (Fig. 9). The main preliminary conclusions are summarised below:

- All the tested membranes (1C-, 19C-, 151C-, 163C-) attained a COD retention of over 80 %, regardless of the membrane area.
- The EC rejection was in the range of 30–80 %; the desired limit of 400 µS/cm was not reached due to the inability to completely retain monovalent ions.
- With an increasing feed pressure, the permeability decreases, while the micropollutants are removed more efficiently.
- The NF showed high efficiency in retaining heavy metals (Cu^{2+}, Zn^{2+}, Pb^{2+}), achieving up to 99 % removal. All the required recycling limits were reached.
- As the CHMS laundry company receives carpets and towels from different industries and countries, it is expected that the wastewater will vary from time to time, influencing the membrane performance.

In continuation of the project, installation of a pilot plant at CHMS is planned. At this moment, the pilot plant is in construction by the project partner E.S.C.H. According to the project schedule, this pilot plant will be installed at CHMS during 2022.

The project goal is to present a demonstrated technology for recycling of laundry wash waters, coming from different sources and different levels of contamination, on time, by the end of 2024.

Fig. 9
Laundry wastewater sample – feed (l.), produced permeate (r.)

Acknowledgments go to the [German] Federal Ministry of Education and Research BMBF for funding.