2009 summer

Membrane News Twente

News magazine of the Membrane Technology Group

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Solid State Ionics

Research within this interdisciplinary field of material science is one of the main activities of the Inorganic Membrane group. It encompasses the study of fast ionic and mixed ionic-electronic transport in solids and associated interfacial/electrode reactions. Related material characteristics are also investigated to address and understand the basic transport properties, or how these affect the long-term stability of the targeted materials. Essential to the research is the development of a fundamental understanding of the relationships between material characteristics, such as crystal structure, composition, defect chemistry and microstructure, and the material properties related to diffusion of ionic and electronic species. The research relates to a number of important technological applications, such as oxygen transport membranes, oxygen storage materials, solid oxide fuel cells (SOFC's, Figure 1), solid acid fuel cells (SAFC's) and high-temperature proton-conducting fuel cells (PCFC's), oxygen and NOx sensors.

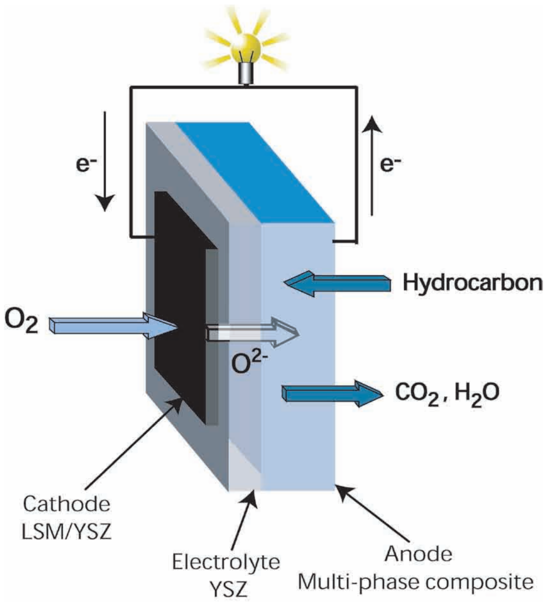


Figure 1: Operating principle of a solid oxide fuel cell.

Mixed conducting ceramic membranes

Ceramic membranes made from mixed ionic-electronic conducting oxides (MIEC's) selectively separate oxygen from air at elevated temperatures, typically higher than 700°C. By imposing an oxygen partial pressure gradient across the MIEC membrane, oxygen permeates from the high to the low pressure side of the membrane. Owing to the ability that both oxygen anions and electrons are mobile, the MIEC membrane operates without the need of electrodes and external circuitry, by contrast with operation using the solid oxide fuel cell or oxygen pump.

The mixed conducting ceramic membranes, also known as oxygen transport or ion transport membranes, are being developed for numerous industrial processes that need a continuous supply of oxygen, for example, for the production

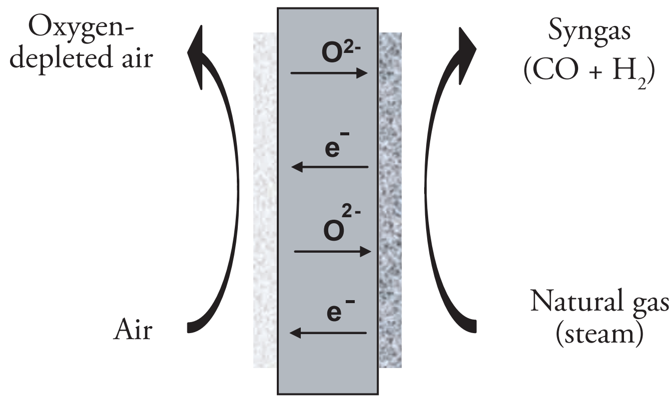


Figure 2: Operating principle of a membrane reactor incorporating a mixed conducting membrane for the partial oxidation of methane to syngas.

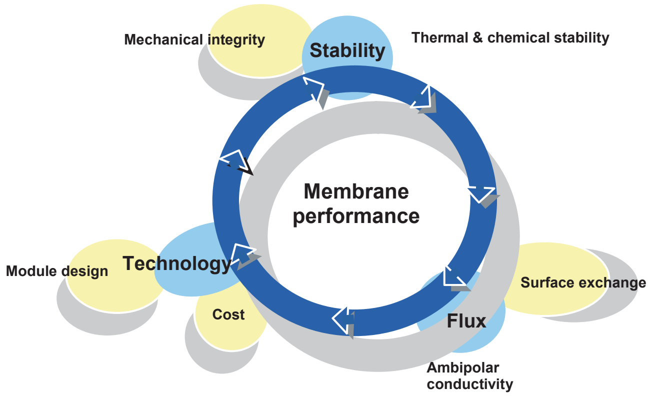


Figure 3: Schematic diagram expressing the interplay and synergy between flux, stability and technology on membrane performance.

of syngas by partial oxidation of methane (Figure 2). Use of the membranes in oxyfuel combustion (e.g., combustion of fuel using pure oxygen rather than air as the oxidant) in power plants is an enabling technology for the capture and subsequent sequestration of CO_2. However, a major barrier to technological application is the high temperature of operation to obtain acceptable oxygen fluxes. Many material problems still need to be solved, among which the thermal, chemical and mechanical stability to improve the life time and reliability of operation of the membranes (Figure 3).

Solid acid fuel cell

Inorganic solid acids like $CsHSO_4$ and CsH_2PO_4 exhibiting high proton conductivity have potential for use as electrolyte in intermediate temperature fuel cells. Unlike hydrated sulphonated polymers such as Nafion®, no water molecules are required to facilitate proton transport in the solid acids, eliminating the need for continuous humidification of reactant gases.

Proton conductivity in the solid acids arises upon a structural phase transformation. The transition, often referred to as superprotonic phase transition, creates dynamical disorder in the H-bonded XO_4 network (where X= S, Se, P), enabling fast transport of protons mediated by rapid reorientations of the XO_4 tetrahedra (Grotthuss mechanism).

Recent identification was made in the Inorganic Membrane group of high proton conductivity at ~140 °C in the solid acid $KH(PO_3H)$. Unlike sulphate and selenate solid acid electrolytes, $KH(PO_3H)$ can be operated in both oxidizing and reducing atmospheres. Slight humidification of the gases in liquid water at room temperature, with an equivalent p_{H2O} of 0.02 atm, is sufficient to prevent impairment of the proton conductivity due to dehydration of the solid acid. Figure 4 shows the conductivity of a ~10 μm thick nano-sized SiO_2-dispersion strengthened $KH(PO_3H)$ membrane.

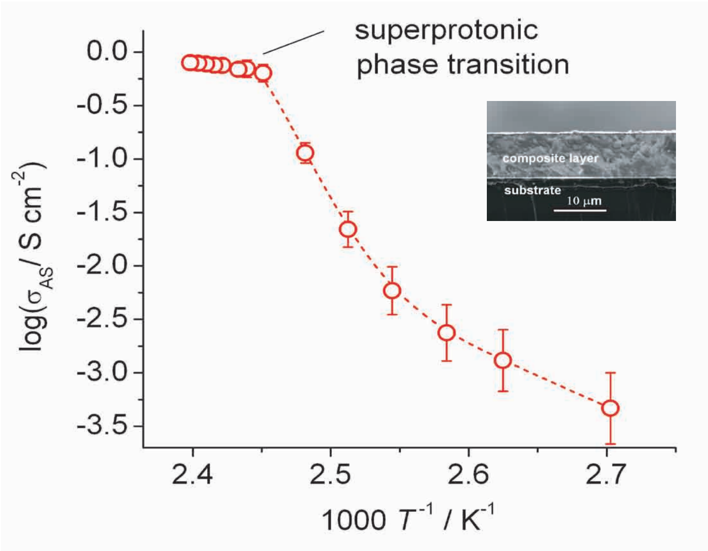


Figure 4: SEM image and area specific conductivity (σ_{AS}) of a $KH(PO_3H)$-SiO_2 thin film (~10 μm).

In a joint cooperation with researchers from the Transducer Science and Technology group of the University of Twente work is ongoing to incorporate the novel solid acid electrolyte in a micromachined solid acid fuel cell for mobile applications.

For more information, please contact Dr. Henny J.M. Bouwmeester (h.j.m.bouwmeester@tnw.utwente.nl; phone: + 31 (0) 53 489 2202

University of Twente hosts ICOM 2011

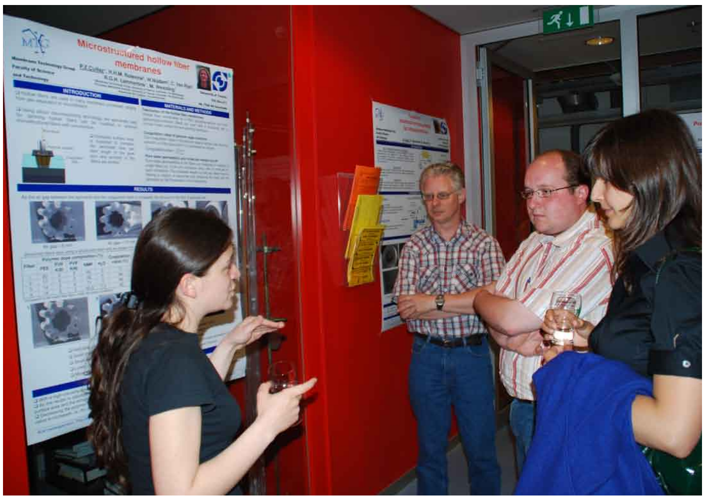
In 2011 the ICOM (International Conference on Membranes and Membrane Processes) will be hosted by the Membrane Technology Group of the University of Twente. ICOM is the largest international conference for the exchange of fundamental and applied knowledge and research on membrane science and technology. The event will take place in Amsterdam from July 23 till July 29, 2011.

Twente Membrane Day

On May 29th 2009 the Membrane Technology Group (MTG) hosted a half day symposium at the University of Twente. Aim of this symposium was to provide an update on current and future membrane research activities in Twente, to show the MTG laboratory facilities in the new Meander building, and to provide an informal ambiance for dialogue with colleagues in the field. The symposium, Twente Membrane Day, has been organized with irregular intervals since 1998. It is free of charge and open to everybody that is interested in membranes. This year, just over hundred people attended, approximately half of which came from outside the University.

The symposium started with four lectures, each linked to one of the four different entities within MTG. In the first lecture Matthias Wessling presented research highlights of the chair Membrane Science and Technology, emphasizing on micro-structuring of membranes and on the accommodation of functional particles in porous polymer hosts. Antoine Kemperman provided an overview of current advances in research on membrane fouling carried out within the chair Membrane Process Technology. Challenges faced in the Inorganic Membranes group were reviewed by Henny Bouwmeester, and finally Zandrie Borneman gave an outline of the possibilities for contract research within the European Membrane Institute (EMI). The lectures were followed by a guided tour through the MTG laboratories, in turn followed by a poster session and drinks.

Appreciating the positive response of the participants, plans for the next Twente Membrane Day are already in progress. For those who can not wait that long, you are more than welcome to visit us at a time that is more convenient for you.



Reverse electrodialysis for sustainable energy generation

Reverse electrodialysis (RED) (Figure 1) is a promising and potentially attractive technology for the generation of sustainable energy from the mixing of salt and fresh water. It uses the free energy of mixing of two solutions of different salinity (e.g. river and sea water) to generate power.

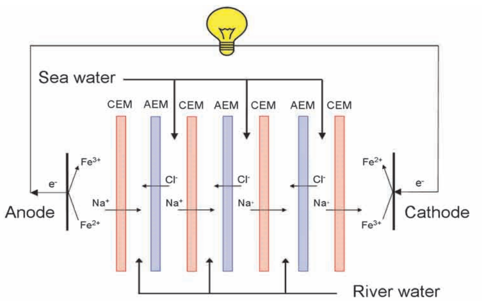


Figure 1: Schematic representation of reverse electrodialysis.

In RED, a concentrated salt solution and a less concentrated salt solution are brought into contact through an alternating series of anion exchange membranes (AEM) and cation exchange membranes (CEM). Anion exchange membranes contain fixed positive charges which allow anions to permeate through the AEM towards the anode and cation exchange membranes contain fixed negative charges which allow cations to be transported through the CEM towards the cathode. At the electrodes a redox couple is used to mitigate the transfer of electrons. The ion exchange membranes are one of the key elements in the RED process as they, together with ion transport phenomena and concentration polarization effects occurring at the membrane-solution interface determine to a large extent the net power output of the system. This PhD thesis investigates the design, optimization and potential of the reverse electrodialysis process, with a special focus on membrane properties, mass transfer of the ionic species and concentration polarization.

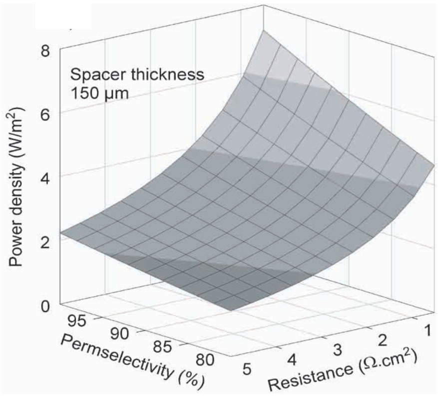


Figure 2: Relationship between power density, membrane permselectivity and membrane cell pair resistance for a membrane pair with a 150 μm thick spacers. Model calculations are based on sea water (0.5M NaCl) as concentrated salt solution and river water (0.05MNaCl) as diluted stream(T=25°C).

Characterization and evaluation of commercially available ion exchange membranes shows that the membrane resistance should be as low as possible, while the membrane selectivity is of minor importance. Based on the results, the best benchmarked commercially available anion exchange membranes reach a power density of more than 5 W/m^2 whereas the best cation exchange membranes show a theoretical power density of more than 4 W/m^2. According to membrane model calculations power densities higher than 6 W/m^2 can be obtained by using thin spacers and tailor made membranes with low membrane resistance and high permselectivity especially designed for reverse electrodialysis (Figure 2).

In most literature, it is assumed that the membrane resistance is independent of the concentration and corresponds to the value determined in a standard characterization solution (0.5M NaCl). In RED however, much lower salt concentrations are used and our experimental work reveals that especially at low solution concentrations (< 0.1 M NaCl) the resistance of ion exchange membranes strongly depends on the salt solution concentration and a very strong increase in membrane resistance with decreasing concentration was observed (Figure 3).

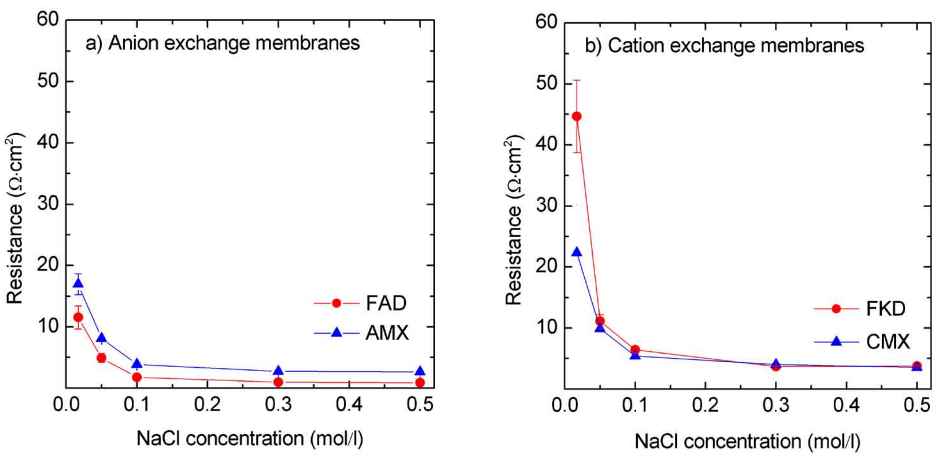


Figure 3: Membrane resistance as a function of NaCl solution concentration for a) anion exchange membranes and b) cation exchange membranes. Values are obtained from measurements under direct current conditions.

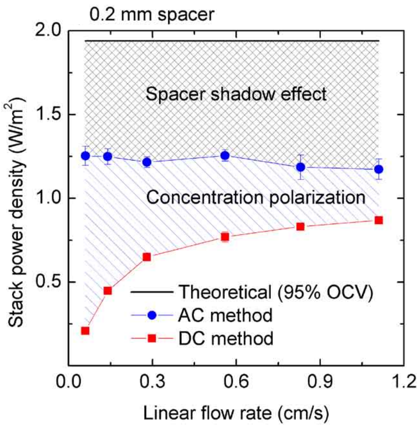


Figure 4: Stack power density as a function of the linear solution flow rate for 0.2 mm spacers. All measurements performed at 25 °C

The underlying reason for the increase in membrane resistance at lower salt concentrations is further investigated using direct and alternating current characterization methods. As the direct current characterization method is unable to distinguish between the membrane resistance and the contributions of the electrical double layer and concentration polarization phenomena at the membrane-solution interface, we used electrochemical impedance spectroscopy (EIS), which uses an alternating current, to analyze resistance phenomena in ion exchange membranes under different hydrodynamic conditions and at different temperatures. The results clearly show that the increase in membrane resistance with decreasing concentration as measured in direct current experiments is the consequence of the strong increase in resistance of the diffusion boundary layer with decreasing concentration. This also explains the strong effect of the flow rate on the resistance. At higher concentrations, the pure membrane resistance starts to dominate the resistance as measured by direct current measurements, although also the diffusion boundary layer resistance still plays a considerable role.

Theoretically, salinity gradient energy has a global potential of 2.6 TW as renewable energy source. Although the theoretical potential is high, the practical power output obtained is limited yet due to concentration polarization phenomena and spacer shadow effects. We combined theoretical calculations with direct current and alternating current experimental stack characterization methods to quantify the contribution of concentration polarization phenomena, spacer shadow effects and stack resistance in RED under different hydrodynamic conditions in a temperature range from 10 to 40 °C (Figure 4).

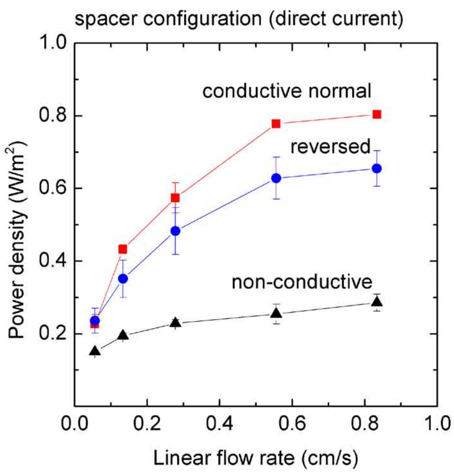


Figure 5: Practically obtainable stack power density as a function of linear solution flowrate for a 0.32 mm spacer. All measurements performed at 25°C.

Increase in temperature from 10 to 40°C results in a strong increase in stack power density. Concentration polarization phenomena play an important role and their influence can be minimized by optimal stack hydrodynamics. Improved spacer designs and new spacer concepts offer extensive room to reduce the spacer shadow effect and to further increase the practical power output.

The spacer shadow effect in RED occurs when non-conductive spacers are used that block the ionic transport in the stack thus reducing the area effectively available for ionic transport. This has a high impact on the net power output. To overcome this effect, the use of ion conductive spacers is investigated (Figure 5).The use of such ion conductive spacers led to a decrease in stack resistance by a factor 2 and an increase in power density of a factor 3 compared to the use of non-conductive spacers made of PVC with the same open area and shape.

This work not only evaluates the challenges in RED and assesses important research directions for further development and scale-up that need to be addressed before reverse electrodialysis can be referred to as a full scale and mature technology, it also shows the strong potential of RED as source for sustainable energy generation from salinity gradients.

For more information please contact Dr. Kitty Nijmeijer (d.c.nijmeijer@utwente.nl; phone: + 31 (0)53 489 4185).

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Hollow fiber membrane coating for CO_2 capture applications

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Nicolas Hengl is a new Post Doc in the Membrane Technology Group. He finished a technical degree in Chemical Engineering in Toulouse at the University Paul Sabatier France and he followed Physical Chemistry studies in Montpellier where he obtained his Msc. In November 2005, he started his PhD at the European Membrane Institute in Montpellier, France. His research focused on the simulation and the conception of a new membrane contactor using metallic membranes for the concentration of thermal sensitive products. The goal of his post doctoral project is the study of hollow fiber membrane coating and its scale up for CO_2 capture applications.

DSTI researcher in Membrane Technology Group

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Jumeng worked as DSTI researcher in the Membrane Technology Group from April 2009. The main focus of his research is nanofiltration under extreme conditions. Jumeng received his PhD degree in chemistry in 2003 from Zhejiang University where he worked on hollow fiber membrane contactors. Subsequently, he has worked on a number of projects covering various topics, including catalytic membrane reactors, freeze-casting synthesis of ceramic monoliths, gas separation membranes, and robust pervaporation membranes.

Membrane reactors for syngas and hydrogen production

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Tan was born in Vietnam. He obtained his BSc. degree on Silicate Materials Technology in HoChiMinh-city University of Technology in Vietnam. After then, he pursued Master course in Sungkyunkwan University in South Korea and obtained his Msc. degree in August 2008. He studied the fabrication processes and properties of porous reaction-bonded silicon carbide for hot gas filters. He joined the Membrane Technology Group as a PhD student in April 2009. His research focuses on the synthesis and performance of single-phase perovskite membrane and/or dual-phase composite membrane reactors for the production of syngas and/or pure hydrogen.

New technician in Inorganic Membranes Group

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On 1st of May 2009, Frank started as a technician within the Inorganic Membrane group. Main activities of Frank include building and updating of dedicated experimental set-ups. Being born and raised in Enschede, Frank is a real "tukker". After military service he started as a mechanic/toolmaker at the R&D department of a family company. Subsequently, Frank became mechanic mechatronics for the company Power Packer where he built prototype convertible rooftops and hydraulic steering. After 4 years he found a new challenge at the department for nanotechnology of Philips Machinefabriek, building modules for the semiconductor industry. He stayed Philips for about 12 years and obtained a diversity of skills, for example related to tubing and vacuum technology, that are very valuable for his present function.

Introducing...

Platform for ligand immobilization focused on proteins

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Dr. Kali Kishore Reddy Tetala, was born on 5th August 1979 in India. He graduated in 2001 with Master of Science (M.Sc) in Organic Chemistry from Andhra University, India. Later he moved to Southampton University for his Master of Philosophy degree (M.Phil) in organic chemistry with project entitled “Palladium catalysed synthesis of amindines and imidates”. In 2004, he started his PhD work at Wageningen University in the project entitled “Microfluidic devices for sample clean-up and screening of biological samples”. In 2008, he started to work with DSTI as a researcher for the project entitled “New separation principles for oligosacharides based on molecular affinity”. Recently he started as a researcher at the membrane technology group. The aim of the project is to develop “Mixed Matrix membranes (MMM)” consisting of functional particles dispersed in a porous matrix. This allows easy access to the functional particle for high throughput recovery of proteins from product streams.

Mixed matrix membranes for toxin removal from blood

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Marlon Tijink studied Biomedical Sciences at the Radboud University in Nijmegen. During her MSc; she studied effects of benzene metabolites on chromosome segregation and spindle positioning at the Utrecht University. At the end of her MSc she investigated the functional impact of diet-induced obesity on the uterus at King’s College London. She started her PhD project entitled ‘Mixed matrix membranes for toxin removal from blood’ in the Membrane Technology Group in January 2009. Absorptive particles are incorporated into porous membranes to combine filtration and adsorption of several types of toxins in one step. The project is financially supported by the Dutch Kidney Foundation (Nierstichting) and it is part of its initiative for development of a wearable filtration adsorption membrane device for continuous and improved blood cleansing in the treatment of end-stage renal disease.

MTG Study Tour

From Sunday May 10th till Wednesday May 13th the Membrane Science & Technology group went on a study tour to Germany. The program started on Monday with a visit to the 29th ACHEMA, the world’s largest international exhibition-congress on Chemical Engineering, in Frankfurt am Main. Tuesday the program included a visit to Gambro Dialysatoren GmbH in Hechingen were we visited the production location of dialyzers. The afternoon was reserved for social-cultural time with a visit to the Atomkeller-museum in Haigerloch and spare time to explore this compact and beautiful city. The study tour ended Tuesday evening with a dinner at Burg Hohenzollern on top of the 855 meter high Zollernberg and an evening/night with social drinks and laughter. Late Wednesday afternoon the group returned in Enschede exhausted, but satisfied.

Design strategies for Tissue Engineering Scaffolds

Tissue engineering (TE) is an interdisciplinary field applying the principles and methods of engineering and life sciences to fundamentally understand and develop biological substitutes to restore, maintain or improve tissue functions. Basically, TE attempts to mimic the function of natural tissue using a patient's own cells, as illustrated in Figure 1. Constructing a tissue engineered replacement in vitro (outside the body) is considered an excellent alternative to direct transplantation of donor organs by overcoming many difficulties as insufficient donor organs, pathogen transmission and rejection of the donor organ.

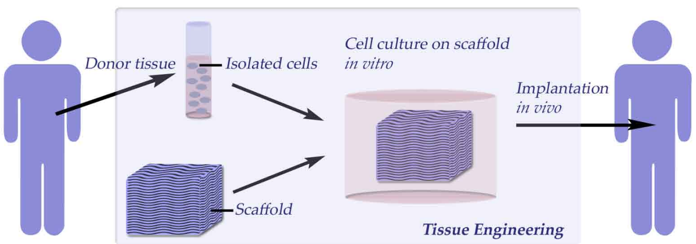

Figure 1. Schematic illustration of the tissue engineering principle.

In vivo, the cells are surrounded by the extra cellular matrix (ECM), which is a meshwork-like substance that supports cell attachment and promotes cell proliferation. Within the TE concept, a scaffold is used as artificial ECM. The scaffold should allow tissue formation in 3D by good support of cell attachment, proliferation, as well as enabling sufficient nutrient supply to the cells and waste elimination from the cells. Besides, introduction of a specifically designed surface architecture can help restoring tissue organization as isolated cells cultured in vitro tend to lose their in vivo organization.

This thesis focuses on various aspects involved in scaffold design and the interaction of cells with the scaffolds. The ultimate goal is to design a scaffold that supports functional tissue formation, resembling in vivo tissue organization, combined with good nutrient supply to the cells. Our concept is based on developing 3D multi-layer scaffolds consisting of porous micropatterned sheets using an one-step fabrication method incorporating both porosity and surface topography, called phase separation micromolding (PSμM). Figure 2 illustrates the multi-layer concept, where cells grow within micropatterned channels giving a clear direction to the cells inducing cell organization. Subsequent stacking of these sheets results in a 3D scaffold where the microchannels also provide space for perfusion of nutrients throughout the complete scaffold. The porosity of the individual single layers enables diffusion of nutrients and signalling factors between the layers.

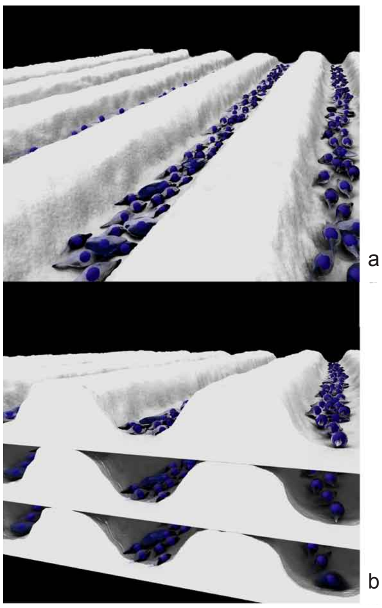

Figure 2. Schematic illustration of the 3D multi-layer scaffold concept

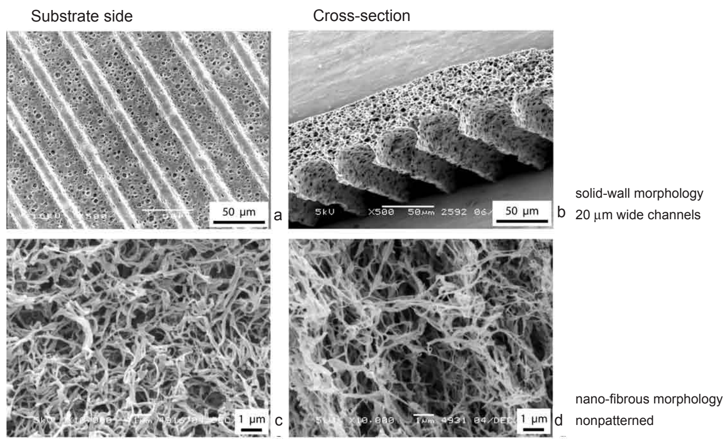

Figure 3. SEM images of PLLA sheets comprising (a, b) a solid-wall morphology combined with 20 μm wide channels and (c, d) a nano-fibrous morphology.

This thesis aims at acquiring insight into the role of scaffold design and how to affect cell-scaffold interactions at various levels, and comprises three main topics: material properties, surface characteristics and 3D architecture.

First, an overview is presented of commonly applied biomaterials as well as fabrication techniques to process these materials into scaffolds, which in contrast to PSμM generally only incorporate or porosity or surface topography. Subsequently, the understanding and optimization of the PSμM processing conditions are considered to obtain the micropatterned porous scaffold sheets of distinct polymers varying largely in material properties, of which poly(L-lactic acid) (PLLA) is the most commonly applied. Adapting the PSμM process parameters yields micropatterned sheets expressing a wide variety of porosities and morphologies. Even micropatterned PLLA sheets comprising nano-fibers of 50-500 nm, similar to the dimensions and morphology of the fibers making up the vast majority of the natural ECM matrix, is achieved. Figure 3 depicts SEM images of various PLLA sheets.

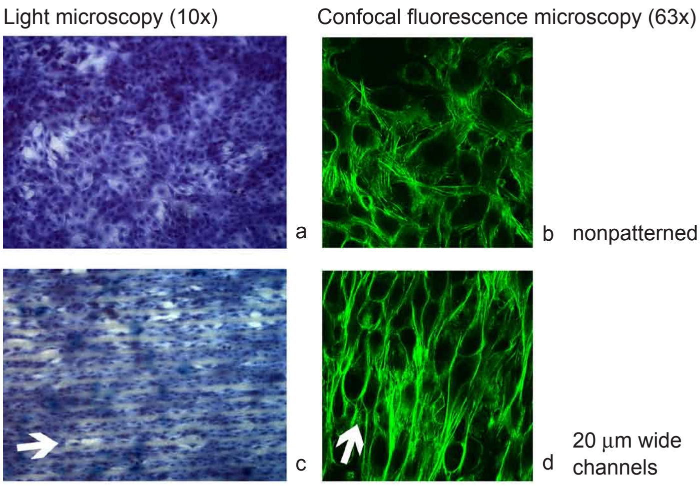

Figure 4. C2C12 cells cultured on PLLA sheets without (a, b) and with (c, d) microchannels. Cells are stained with methylene blue for light microscopy (a, c) or phalloidin cytoskeleton staining for fluorescence microscopy (b, d). The arrows indicate the channel direction.

With respect to the cell-material interactions, the results reveal clear organization of the cells for distinct surface topographies, independently of the scaffold material. Figure 4 illustrates the alignment of C2C12 mouse pre-myoblast cells on sheets featuring microchannels versus a random orientation of the cells cultured on nonpatterned sheets. Interestingly, when the cells grow multi-layers and the cells overgrow the microchannels, they still maintain their organization. Besides the effect on cell organization, surface topography also affects other cell behavioural aspects as cell attachment, morphology and proliferation. A detailed study presented in this thesis clearly relates the influence of scaffold material and micropatterning to surface wettability and protein adsorption, and in turn their effect on cell attachment, cell proliferation and cell morphology. Furthermore, we present a new concept based on high-throughput screening of a large surface characteristics library to enable selection of surface characteristics that elicit an appropriate bio-active response required for a specific application.

Finally, the transport of nutrients throughout the 3D multi-layer scaffold is studied under both static as well as dynamic (perfusion) conditions. Nutrient perfusion through the microchannels of the scaffold clearly revealed improved cell viability and proliferation at the distinct layers showing the excellent potential of the multi-layer 3D scaffolds comprised of micropatterned porous sheets as proposed in this thesis.

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Bernke Papenburg defended her thesis on June 19th, 2009. From April till mid August 2009, she will work a PostDoc researcher within the same project. On September 2009, Bernke will start a new line of PostDoc research at the department of Biomedical Engineering at the Tufts University, Medford, MA, USA under the supervision of Prof. D. Kaplan. For more information please contact Dr. Dimitris Stamatialis (d.stamatialis@utwente.nl; phone: +31 (0) 53 489 46 75.

Membrane Runners participate in Batavierenrace

The Membrane Technology Group maintained its tradition to participate in the annual runners event, the Batavierenrace, which was organized on April 24 and 25, 2009.

The Batavierenrace is a 185-kilometer relay race from Nijmegen to Enschede, divided into 25 stages (17 men's and 8 women's stages) varying from 3.4 km to 11.9 km. More than 7,500 students (300 teams) participate in this race. In 1972, a couple of students from University of Nijmegen came back from the SOLA relay run in Sweden with so much enthusiasm, that they decided to organize a similar event in the Netherlands. These students inspired academics and they chose the route of the Batavians, who sailed down the Rhine in 50 B.C. The first race followed the original route to Rotterdam, but the route was later (from 1974 onwards) redirected to Enschede because of infrastructural problems. More information you can find at: http://www.batavierenrace.nl/

The group once again registered under its trademark name "Membrane Runners". The membrane scientist gave their best and performed quite well on all the 25 stages. Although the overall ranking of the group was 210, we ranked 17th the 7.6 km distance achieved by our Postdoc Dr. Nicolas Hengl, who completed the distance at an average speed of 15.02 km/hr. Our fastest female runner was Uli Adams.

The Membrane Runners will keep running in coming years and continue to show their competences in a field outside their daily research.

To round up the success of this event the Membrane Technology group hosted a barbecue at Louis van de Ham's place, followed by the biggest student party in Europe on the Twente University campus.

We would like to thank all our sponsors for supporting the membrane technology group, since without their contribution this event might not have been possible. The Membrane Runners would like to express their gratitude to Walter van der Meer from Vitens, Sybrand Metz from Wetsus (Research Institute for Sustainable Water Technology), Rob Kreiter from ECN and Zandrie Borneman and Antoine Kemperman from the European Membrane Institute.

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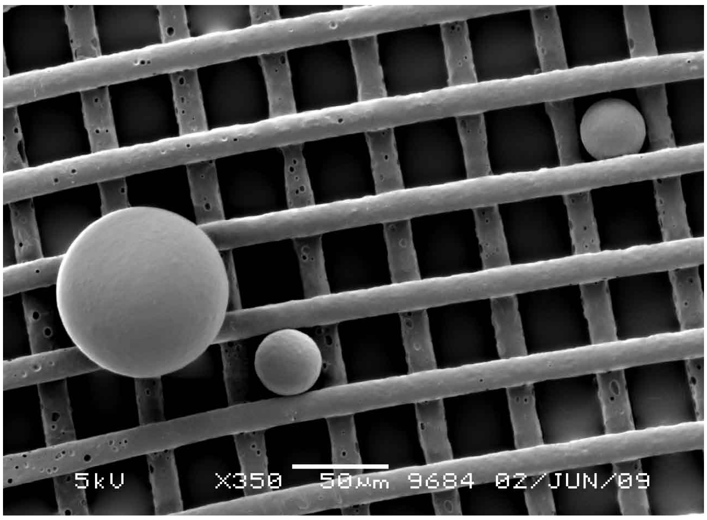
Theses

Piotr Dlugolecki
Fall, 2009 (specific date to be determined)
Reverse Electrodialysis for sustainable energy generation

Matias Bikel
December 18, 2009
Phase Separation μ-Molding: Fundamental Aspects



Filtration of polymeric particles through a polymeric mesh made by Vapor Induced Phase Separation MicroFabrication using PDMS molds. Picture by Matias Bikel

MNT- Information

Membrane News Twente is published two times per year. The aim is to inform the membrane community about the activities of the Membrane Technology Group.

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Internet & MNT archive
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Circulation
~500