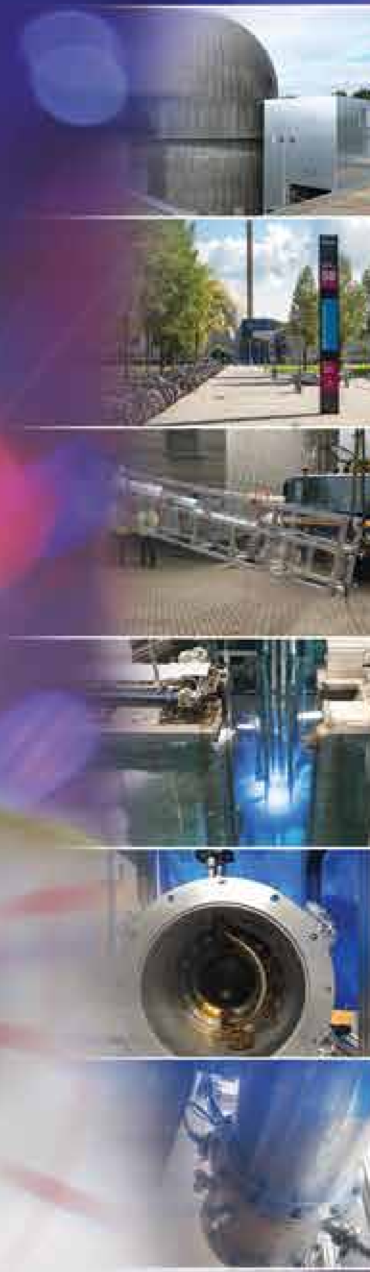
2019

OYSTER

Annual Report

Final preparations for the cold neutron source



TU Delft Delft University of Technology

REACTOR INSTITUTE DELFT

Foreword

This annual report chronicles the last stages of the OYSTER programme (*Optimized Yield - for Science, Technology and Education - of Radiation*), which was initiated to improve and expand the RID research infrastructure at TU Delft. OYSTER will provide us with powerful new tools to address educational, scientific and societal challenges in domains such as materials, health and energy. In 2019, the research reactor was shut down to prepare for the culmination of the project: the installation of the cold neutron source.

First, in February 2019, the eagerly awaited mock-up of the cold neutron source arrived from South Korea. We used the mock-up as a test facility to verify correct functioning before the actual cold source will be manufactured and ultimately installed in the reactor pool. The mock-up was also used to train our team.

At the same time, we pushed on with instrument development, focusing on instruments that 'view' the cold source. Connected to the operation of these instruments, instrument scientists were hired for the daily running of the ROG and SANS instruments. The neutron reflectometer ROG will contribute to addressing many scientific challenges, such as the development of hydrogen sensors and drug-delivery systems. SANS is a powerful scattering technique to support the development of, for example, solar cells and nanocomposites.

We also started to build a second flexible irradiation facility with a pneumatic station. Check out the interview with Quirem Medical in this report. The company uses our facility to produce radioactive microspheres for the radiation treatment of cancer patients.

As I write this foreword, we are in the midst of the COVID-19 pandemic, and it is unsure to what extent the OYSTER planning will be affected. In 2019 we experienced some delays due to subcontractor issues, but if everything goes ahead as planned, the OYSTER project will be concluded in late 2020.

We are really looking forward to the moment the cold neutron source will be placed in the heart of the research reactor and connected to the cooling equipment in the CNS-Utility building. The neutrons from the reactor will then be cooled with liquid hydrogen to an extremely low temperature of minus 253 degrees Celsius. RID's instruments will provide results up to a hundred times better than now, a goal we have been working towards for years. We can't wait to explore, exploit and share our new and improved facilities with researchers around the world.

I hope you will enjoy reading this report.

Prof. dr. Bert Wolterbeek
Director of the Reactor Institute Delft

Colophon

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OYSTER in short

The Reactor Institute Delft (RID), part of Delft University of Technology (TU Delft), is a nuclear knowledge centre. It operates a research reactor, irradiation facilities and radiation-based research instruments. The OYSTER programme (Optimized Yield - for Science, Technology and Education - of Radiation) has been granted in 2012 to improve and expand the RID infrastructure (reactor, instruments, facilities). This will enable current and future educational, scientific and societal questions to be better addressed. In conjunction with the Department of Radiation, Science & Technology (RST) of the Faculty of Applied Sciences, RID accommodates resident and visiting scientists and other users from a variety of (scientific) disciplines. RID educates students, professionals and scientists, and serves as an independent source of information for society on radiation and nuclear-related issues.

OYSTER

Main goals of OYSTER

RID is active in the field of neutron, positron, reactor and radiochemistry science. Here, OYSTER has five main goals:

1. To strengthen RID's national coordinating role.
2. To establish RID's European role in research and training.
3. To stimulate ground-breaking innovations.
4. To create a home base for neutron scattering in the Netherlands and secure Dutch collaboration with major international neutron sources, specifically the European Spallation Source (ESS) in Lund, Sweden.
5. To sustain RID's leading role in the knowledge and use of world-class instruments, in the development of new routes for radioisotope production and in Instrumental Neutron Activation Analysis (INAA).

Technological objectives of OYSTER

OYSTER is an ambitious programme of technological improvements and additions to the RID infrastructure:

- The installation of a Cold Neutron Source (CNS), cooling neutrons from room temperature to -250°C, to increase the intensity of low-energy neutrons by more than an order of magnitude and improve the sensitivity of existing instruments.
- The design and construction of new research instruments.
- The (re) design and construction of (new) irradiation facilities, to allow the development of production of radioisotopes with unprecedented purity and to boost the sensitivity and opportunities for research with isotopically enriched stable isotopes.
- The design and installation of a miniature hot-cell/decanning facility for submerged access of irradiated samples from the irradiation facilities, to allow innovative production methods of (medical) radioisotopes and studies of radiation damage effects.
- The design of irradiation facilities positioned in the tangential beam tube of the reactor, to undertake research into alternative production methods for e.g. molybdenum-99 (^{99}Mo).

Participation in large international and national collaborations

OYSTER will be instrumental in securing or strengthening the role of the RID in various collaborations. For example, the Dutch contribution to the pre-construction phase of the European Spallation Source (ESS, an international collaborative facility for materials research using neutron scattering techniques) in Lund, Sweden, is partly financed through OYSTER. Scientists from Delft work together with ESS scientists in order to develop novel instrument concepts for the ESS.

RID participates in the R&D programme of Holland Proton Therapy Centre (HollandPTC), dedicated to innovative radiation treatment of cancer using proton beams. This programme is a collaboration of TU Delft, Leiden University Medical Center (LUMC) and Erasmus University Medical Centre Rotterdam (Erasmus MC). Located on the RID premises, HollandPTC began patient treatments in September 2018.

OYSTER research also strengthens the role of RID in supplying innovative ideas towards the envisioned PALLAS reactor, which is to become one of the world's leading production sites for medical isotopes.

Photo: Campus design of the European Spallation Source (ESS) in Lund, Sweden. Photo: ESS.

Photo: Holland Proton Therapy Centre (HollandPTC) on the RID premises.

The OYSTER-initiated new irradiation facilities also enhance RID's position in the Dutch Isotope Valley (DIVA). This is an R&D collaboration set up between URENCO, RID and NRG/PALLAS towards developing, engineering and producing the best-possible medical isotopes for clinical use (both diagnosis and therapy).

Finally, OYSTER underlines the role of RID as an IAEA Collaborating Centre by demonstrating the many innovative scientific opportunities in the utilisation of a medium-sized university research reactor.

Reactor & Utilities

Overview

A Cold Neutron Source (CNS) will be installed in the reactor pool and is therefore designated as the CNS In-Pool Assembly (IPA). The modification of the reactor deals with the installation of CNS-IPA into the beam-tube between the reactor core and instrument facilities located in the instrument hall. The objective of this modification is to increase the cold neutron flux in order to create better conditions for experiments connected to the neutron beam. To supply the cooling capacity for the proper functioning of the CNS, support equipment was already installed in the brand-new CNS-Utility building which was realised next to the reactor hall.

Reactor modifications

Reactor shutdown

To facilitate the reactor modifications, the reactor was shut down from May 2019 onwards.

Mock-up cold neutron source

In February 2019, the mock-up of the In-Pool Assembly (IPA) arrived from Korea. The mock-up is a model for the main IPA that will be installed in 2020. The mock-up IPA is a two-layer system (hydrogen and vacuum layers) that was installed next to the CNS-Utility building whereas the final IPA is a three-layer system (hydrogen, vacuum and gas blanket layers) that will be installed in the reactor pool. The mock-up was made to test the thermosiphon concept (see "Hydrogen cooling") and to test the helium refrigeration system before the main IPA will be manufactured.

During initial testing, a deterioration of the vacuum level in the mock-up IPA was measured. This was caused by a small hydrogen leak due to a welding defect. As a result of this leakage we temporarily suspended the tests until a team from KHC flew in to repair the mock-up. We then resumed the tests and KHC successfully performed the scheduled thermosiphon test. To prevent reoccurrence of such a welding defect, the design of the affected weld was changed and additional third-party quality control was added to the manufacturing process of the main IPA.

Photo: Testing with the mock-up

The results of the thermosiphon test showed that the mock-up easily reached the temperature required to liquefy hydrogen and that the thermosiphon circulation of hydrogen was well established. The thermosiphon test also indicated that the control concept of the helium refrigeration system needed to be adjusted; this change was successfully implemented and tested.

The RID team also performed neutron void fraction measurements (see "Hydrogen cooling"). The detector for this experiment was developed together with Bonphysics and the Electronic and Mechanical Support Division (DEMO) of TU Delft. It revealed how much liquid hydrogen is present in the moderator chamber of the mock-up, especially while cooling down to or warming up from liquid-hydrogen temperatures. The results confirmed the measurements by the various temperature sensors installed on the mock-up IPA.

Photos: The arrival of the mock-up of the In-Pool Assembly (IPA) from Korea.

During the test and void fraction measurements, the RID reactor operators assisted and operated the CNS utilities. In this way they were trained on the job and got familiar with the installation. This opportunity was also used to write and check the first version of the CNS utility operating instructions.

The mock-up will be removed in February 2020.

HYDROGEN COOLING

The **thermosiphon concept** is a method of passive heat exchange that doesn't require a mechanical pump to circulate the coolant or heating liquid. In the case of the CNS, the coolant is liquid hydrogen, which turns into gaseous hydrogen after heat exchange. Instead of by a pump, the hydrogen starts flowing simply as a result of gravity. Liquid hydrogen is heavier than gaseous hydrogen, so it flows automatically to the moderator cell. Here, the hydrogen evaporates, causing the heat to dissipate due to conduction as well as conventional and nuclear radiation. The vapour then rises and condensation takes place at the heat exchanger, completing the cycle. In practical situations, there may be some (small) gas bubbles carried along with the liquid. This is called 'void' volume. The void fraction designates the ratio between the volume that the void occupies and the total hydrogen volume. While the liquid acts as an excellent coolant to neutrons, the hydrogen vapour has no effect on the temperature of the neutrons.

In-Pool Assembly (IPA)

The fabrication drawings were slightly modified as a result of the lessons learned during the mock-up tests. After approval for manufacturing by RID, the production process was started. Fabrication will be completed in 2020. The IPA will then be installed in the beam tube near the reactor core.

Beam tube modification in the reactor pool

In order to install the Cold Neutron Source (CNS) near the reactor core, a modification of the beam tubes will be necessary. During construction of the research reactor in the 1960s, the beam tubes were designed and built to come close to (but not touch) the reactor core. That way, the neutrons from the nuclear reaction could be directed safely to experiments outside the reactor pool without losing neutron flux due to absorption in the pool water.

The primary modification is a shortening of beam tube R2, which allows for the placement of the moderator chamber of the CNS where the neutrons will be cooled using liquid hydrogen. To access this position, beam tube R1 also needs to be modified. The primary goal of the sub-project 'Beam Tube Modification' (BTM) is to provide space for the CNS assembly. The CNS module and an extension part of the beam tube will be mounted using a quick-disconnect system (QDS) that both preserves the primary water barrier and allows for easy replacement if necessary. This QDS can be remotely opened and closed.

We made much progress towards this goal. KHC, the main contractor for the OYSTER project based in South Korea, completed the design work for the new beam tube components as well as the QDS clamps and seals (through a sub-contract to Technetics, France). The QDS clamp system will replace the current screw-type fastening of the beam tubes that was designed in the 1960s. Fabrication of all components is underway as of December 2019 and the components are expected to arrive in Delft early 2020.

Bilfinger Noell GmbH, Germany (BNG) was selected as a sub-contractor by KHC for the actual modification on-site. Using the designs made by KHC and Technetics, they performed extensive mock-up tests at their worksite in Würzburg, Germany to test their equipment and procedures. Based on these experiences, they have provided updated test plans and procedures. Once the actual components will be available early 2020, a final test run will be performed at their worksite before start of the work in the RID reactor hall.

There were some complications along the way. The procurement of the raw material has been challenging because of the strict requirements and small volumes. This has delayed the start of fabrication. Similarly, the design of the QDS components has taken longer than expected to ensure its functionality as the primary water barrier.

Now the fabrication of the components is nearly complete, and the preparations for the actual installation are also reaching completion, the coming year will be an exciting one. Important checkpoints will be the final test of the components by KHC, the mock-up tests by BNG, and of course the installation of the modified beam tubes themselves in the research reactor.

Reactor hall feedthroughs

To be able to make the connection between the equipment within the CNS-Utilities building on the one hand and the CNS on the other hand, new feedthroughs towards the reactor hall had to be made. The main prerequisite for the reactor hall feedthroughs is that they can be automatically closed off by an isolation valve located on the outside, when needed for safety reasons. Furthermore, it should be possible to verify the leak tightness of the feedthrough locally. In the OYSTER project, these requirements were met by installing new penetration boxes which enclose several process pipes and have electro-pneumatic isolation valves installed on the individual pipes on the outside.

In the feedthrough installation phase, the old box was removed and the two new openings created. During the detailed design of the new reactor hall penetrations, strength calculations on the reactor hall made clear that the temporary situation with three openings did not have enough safety margin for the stability of the reactor hall structure. To maintain sufficient safety margin, extra reinforcement strips were welded to the existing reactor hall. After locally removing the cladding of the reactor hall under asbestos conditions on the outside and removing the chromium (VI) paint under the appropriate conditions on the inside, these reinforcement strips were installed in the final weeks of 2019.

In January 2020, the required openings will be made in the reactor hall by water jetting and the new pre-fabricated penetration boxes will be welded in place. After completing the bridge structure (equipment room) between the CNS-Utility building and the reactor hall, the required pipes and isolation valves can be installed.

Reactor protection system

RID has a reactor protection system in place to help determine possible risks from a myriad of sensor readouts. During the OYSTER period, the current relay-based voting logics of the reactor protection system will be replaced by magnetic core voting logics. This solution was selected for its superior safety and reliability and for the lack of software needed to operate the logics.

The factory acceptance test of the new magnetic core voting logics system for the reactor protection system was performed in June 2019 and witnessed by the Dutch Authority for Nuclear Safety and Radiation Protection (ANVS). After resolving minor issues observed at the factory, the system was shipped to Delft.

Photo: New reactor protection system.

Photo: The reactor hall and the CNS-Utility building.

Wire by wire, the old system was removed to verify the correctness of all the drawings that were used as engineering input. No major discrepancies were observed. After removal of the old system, the new system was installed. Power-up was successful at the first try and an extensive test protocol was followed to come to a Site Acceptance Test of the voting logics.

In 2020, we will replace all solenoids by a more reliable type and configuration matching the output voltage of the new voting logics.

(Sub)contractors

The following executive parties are involved in the realisation of the project-related work packages.

Main contractor

KHC
Consortium of the Korea Atomic Energy Research Institute (KAERI), Hyundai Engineering and Hyundai Engineering & Construction responsible for the CNS IPA realisation

Subcontractors

ROYAL HASKONING DHV
Advising and development total construction.

STRUKTON
Construction company for the civil works related to the Cold Neutron Source (CNS) interface.

DH INDUSTRIES / STIRLING CRYOGENICS
Main supplier of the cryogenic equipment that provides the cooling capacity for the CNS.

KREBER
Main supplier of the vessels inside the CNS Utility Building.

DEMACO
Main supplier of the cryogenic pipelines.

YOKOGAWA
Main supplier of the control systems.

STRUKTON WORKSPHERE
Main supplier of the electrical wire connections.

PROCESS FLOW / DEMACO
Main supplier of interconnecting piping in 2020.

MOOJIN
Supplier of the In-Pool Assembly (IPA) equipment (including CNS and IPA mock-up).

BILFINGER NOELL GMBH
Subcontractor tasked with installing the IPA equipment in the beam tube inside the reactor basin.

STORK
Supplier of the hall protrusion.

External Expert Team

As part of the OYSTER project, many technical discussions have taken place between RID, suppliers and contractors. In order to have sufficient knowledge available, an External Expert Team comprised of external specialists was established in 2014. This team played an important role in the design phase of the project. We thank the members of the External Expert Team for assisting RID in this crucial phase of the OYSTER project. The External Expert Team consists of:

Toni Scheuer

Affiliation: Nuclear Technology Consultant at the TÜV Rheinland Group
Expertise areas: Licensing issues, material and component qualification
Focus within EET: Welding procedures, materials, codes and standards

Stuart Ansell

Affiliation: Neutron Scientist at the European Spallation Source (ESS) in Lund
Expertise area: Cold neutrons equipment design for research reactors
Focus within EET: Optimisation processes neutronics

Stephan Welzel

Affiliation: Chief coordinator of the reactor upgrade at the Helmholtz-Zentrum Berlin
Expertise areas: CNS process technology and operational aspects
Focus within EET: CNS process technology

Robert F. Mudde

Affiliation: Professor of Multiphase Flow, Department Chemical Engineering, TU Delft
Expertise area: Multiphase flows
Focus within EET: Heat and mass transfer, hydrodynamics

Robert Williams

Affiliation: Nuclear Engineer and Cold Neutron Source Team Leader at the National Institute of Standards and Technology (NIST), Gaithersburg, Maryland, (USA)
Expertise areas: Cold neutron sources, calculations and new reactor designs
Focus within EET: CNS performance calculations, operations and safety

Licencing

In the context of Dutch legislation in the area of nuclear safety, RID holds a permit to operate the research reactor and the various instruments. With the OYSTER programme, this permit has to be updated. An important aspect of the OYSTER project concerns the licencing procedures. Since 2013, RID has been working on the licencing procedures and associated review schedules with the relevant regulatory body, the Dutch Authority for Nuclear Safety and Radiation Protection (ANVS).

OYSTER licence

The licence was issued on 29 January 2019 and became fully valid on 15 March 2019, allowing the construction works as part of the OYSTER programme to continue full-speed. Currently there are no further licence issues in this phase of OYSTER.

Ten-yearly safety evaluation

In 2020, a 10-yearly safety evaluation (10EVA) will be carried out, including drawing up a Safety Analysis Report (SAR) for the research reactor. The 10EVA and SAR will serve as foundations for RID to apply for a full licence revision in 2021.

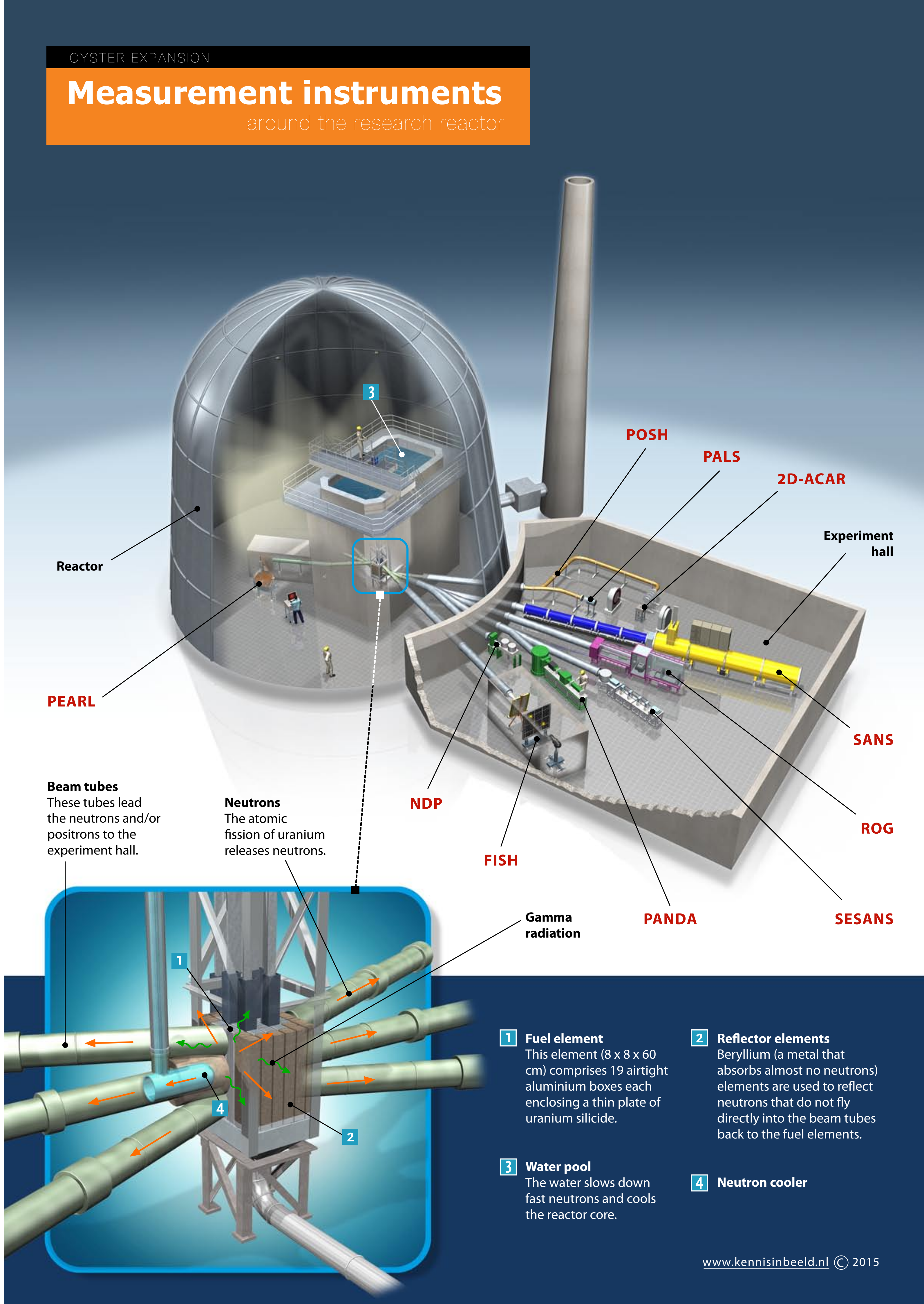
Instruments

As part of OYSTER, RID will develop new or upgrade existing instruments that exploit the (cold) neutron and positron radiation produced by its reactor. The instruments are:

PEARL	the new neutron powder diffractometer
ROG	upgrade and relocation of the time-of-flight neutron reflectometer to a cold beam line
SANS	a new small-angle neutron diffractometer with a dedicated cold beam line
NDP	upgrade of the neutron depth profiling spectrometer
FISH	a new multi-purpose neutron imaging facility
SESANS	upgrade of spin-echo labelled SANS
Mössbauer spectroscopy	upgrade of the existing spectrometer

Overall, the main priority in 2019 was given to the instruments that 'view' the cold source. The Small-Angle Neutron Scattering (SANS) instrument underwent significant detector maintenance programme and the neutron reflectometer (ROG) is undergoing a major upgrade to prepare it for optimal use. The Neutron Depth Profile (NDP) instrument is also being prepared for cold neutron operation with the ordering of a new neutron guide and the design of a new position. The Spin-Echo SANS (SESANS) instrument is being prepared for movement to another beamline to make room for cold neutron imaging that is under design as well. Connected to the operation of these instruments, two persons were hired on instrument scientist/assistant professor (tenure track) positions. They will be responsible for the daily running of the SANS and ROG instruments. For the irradiation facilities, we have officially started to create a second flexible irradiation facility (FlexBeFa) with a pneumatic station.

In the next sections we describe what each of the instruments does or will do, the progress achieved in 2019 and our plans for 2020.



Energy materials

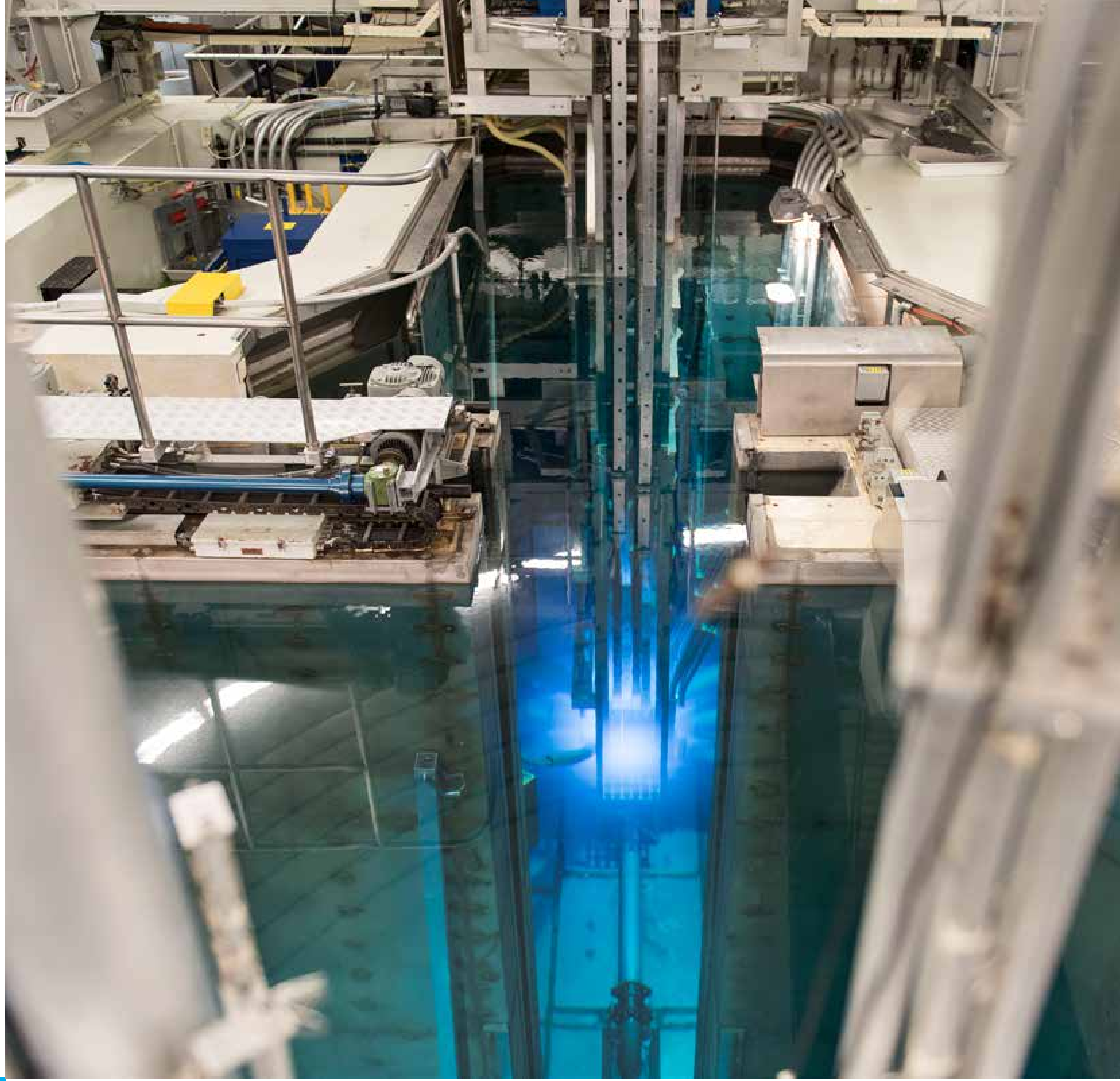
Neutron powder diffractometer - PEARL

The PEARL neutron powder diffractometer was built mainly to determine the crystal structures of energy materials, but its range of applications is much broader. Neutron diffraction and X-ray diffraction complement each other in the sense that neutrons are particularly sensitive to light elements; PEARL is able to distinguish light elements in crystal structures that contain both light and heavy atoms. In addition, magnetic structures and magnetic moments in the crystal can be accurately determined. PEARL is a unique facility within the Netherlands and competitive within Europe.

Progress 2019

During the months before the scheduled reactor stop, follow-up experiments for in-house research were done on batteries, magnetocalorics, skyrmions and nuclear materials. Technical improvements focused on the instrumental background, the further development of a pressure cell for hydrogen gas and sample holders for experiments with the battolyser (a battery with integrated hydrogen conversion and storage capabilities). Within a new research programme in collaboration with the Rijksmuseum Amsterdam, the diffractometer was used to identify the interior of ancient bronzes. Similarly, the benefits of the large penetration depth of neutrons was used on (maritime) archaeological objects, where we could non-invasively probe and identify the original materials in the interior of objects, beyond the corrosion of the exterior. These diffraction experiments were combined with neutron tomography and gamma spectroscopy.

During the scheduled reactor shutdown, the measures that were taken over the last few years to improve data quality (such as background reduction and calibration) were technically improved. We also performed beam-alignment of the sample environment equipment. The number of publications based on PEARL experiments increased with seven.



Photos:
Now that PEARL has a cryostat (left) and a heater (right), measurements can be performed in the temperature range from 1.8K to 800K (-271 to 527 ºC). The design with very thin Vanadium foils ensures that this sample environment has a minimum contribution to the instrumental background in the experimental data.

From hydrogen sensors to drug delivery systems

Neutron reflectometer - ROG

The ROG neutron reflectometer measures the way neutrons are reflected by flat surfaces and interfaces with the aim to provide information about the thickness, composition and roughness of thin films and other layered structures. It can be used to study (stacks of) layers with thicknesses of 5 - 150 nm at resolutions down to 0.2 nm. Neutron reflectometry can be used to non-destructively study both liquid and solid samples in a variety of different experimental conditions. Applications are found in the study of battery materials, metal hydrides for hydrogen sensing and storage applications, membranes, polymers, proteins and drug delivery systems. The ROG neutron reflectometer is currently being upgraded as part of the OYSTER programme. In combination with the installation of the CNS, this upgrade will increase its performance by up to 100 times.

Progress 2019

In 2019, Lars Bannenberg started as Assistant Professor and permanent instrument scientist for the instrument.
We continued our research on thin-film metal hydrides as optical hydrogen sensors resulting in two publications. We also installed, aligned and tested the new optical neutron guide, which will transport neutrons from the reactor to the neutron reflectometer. Finally, we also designed and installed new mechanical components.

Prospects 2020

In 2020, we will complete the upgrade of the neutron reflectometer: we will install new mechanical components including a new versatile sample table, shielding, a new chopper and a position-sensitive detector. This will increase the flexibility, robustness and performance of the reflectometer. We plan to commission the neutron reflectometer with the first neutrons on the sample table.
In addition, we will work towards a new sample environment where we can control the gas pressure (0 – 10 bar) and temperature (max. 350 °C). We will develop an electrochemical cell to study thin-film battery materials and continue our study of thin-film metal hydrides for hydrogen sensing applications.

Photo left:
Instrument scientist
Lars Bannenberg.

Photo right:
New 20 m long neutron guide to transport the neutrons from the cold-neutron source to the neutron reflectometer ROG.

Structures for chemistry, physics, biology and geology

Small-Angle Neutron Scattering instrument - SANS

The Small-Angle Neutron Scattering (SANS) technique allows the measurement of structures on length scales of a few to several hundred nanometres. SANS is a powerful technique of use to disciplines such as chemistry, physics, biology and geology, for example to support the development of solar cells and nanocomposites.
The SANS instrument will take full advantage of the increased cold neutron flux resulting from the OYSTER programme. It will complement the reflectometer (ROG) and the SESANS instrument for large-scale structures

Progress 2019

The instrument awaits first neutrons from the cold source. In preparation for this Assistant Professor Steven Parnell was hired as permanent instrument scientist for the instrument. He has started to establish links within the TU Delft and with the wider scientific community in the Netherlands. In parallel, work on nanocomposites and solar cells was pursued, initially utilizing other neutron sources. In this way, a scientific programme will already be established once the cold source operates at full power.

Prospects 2020

We now await neutrons to begin the commissioning phase of the instrument. In preparation for this, we will test various components: the velocity selector, motors and detector. Dedicated control and analysis software will be written. We are also in the process of upgrading the sample environment with the aim of supporting a range of experimental possibilities as soon as the CNS becomes operational. This includes a new temperature stage and magnet.

Photo:
SANS detector
out of its tank.

Photo: Instrument scientist Steven Parnell.

Battery research

Neutron Depth Profiling - NDP

The Neutron Depth Profiling (NDP) setup is being developed especially for battery research, aiming at high-energy-density batteries for mobile electrical applications. NDP is a powerful technique to detect the litahium ions that are key to such batteries. When a lithium-6 isotope captures a neutron, it sends out two high-energy particles that allow us to study the position and movement of the lithium atom, even in working batteries.

Progress 2019

We previously developed an experimental setup for operando battery research nicknamed "matroesjka". It enables to charge and discharge Li-ion batteries under realistic conditions within the NDP setup. Thereby it allows us to monitor Li-ion transport and irreversible reactions that lead to passivation of Li and hence to capacity fading of batteries.
In the last months of reactor operation before the scheduled shutdown, we performed a large number of operando experiments. We investigated different types of Si anodes, having an almost ten times larger specific Li capacity compared to current graphite anodes, and revealed the inhomogeneity of the lithiation, which we proposed to be one of the critical parameters that determine the cycle life of these electrodes.
We also studied Li metal anodes, having the largest theoretical specific Li capacity, comparing the performance and Li capture mechanisms in different electrolytes.

Finally, operando NDP revealed the redistribution of Li-polysulfides in Li-S cathodes under operando conditions, which is at the origin of the capacity fading of these next-generation high capacity cathodes. Specifically, we studied the preferential adsorption of the Li_yS_x polysulfides on an oxide ($Li_4Ti_5O_{12}$), which can prevent migration of the polysulfides to the anode. These investigations demonstrate that operando NDP offers a powerful new tool in Li-battery research and development, the potential of which we are only beginning to explore.

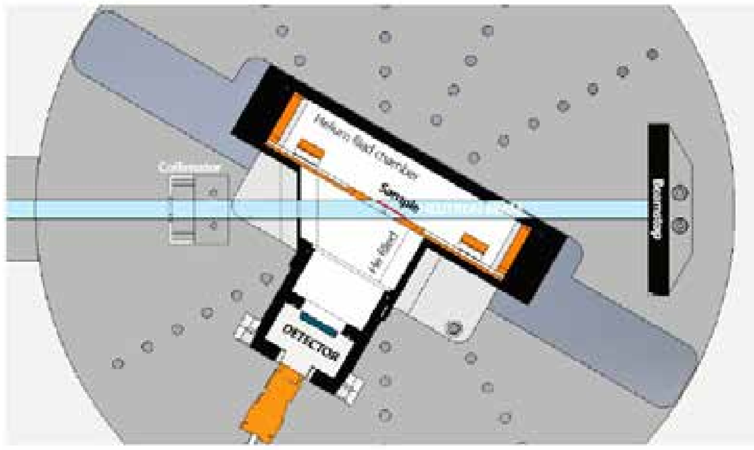


Image: Battery-detector setup "matroesjka"

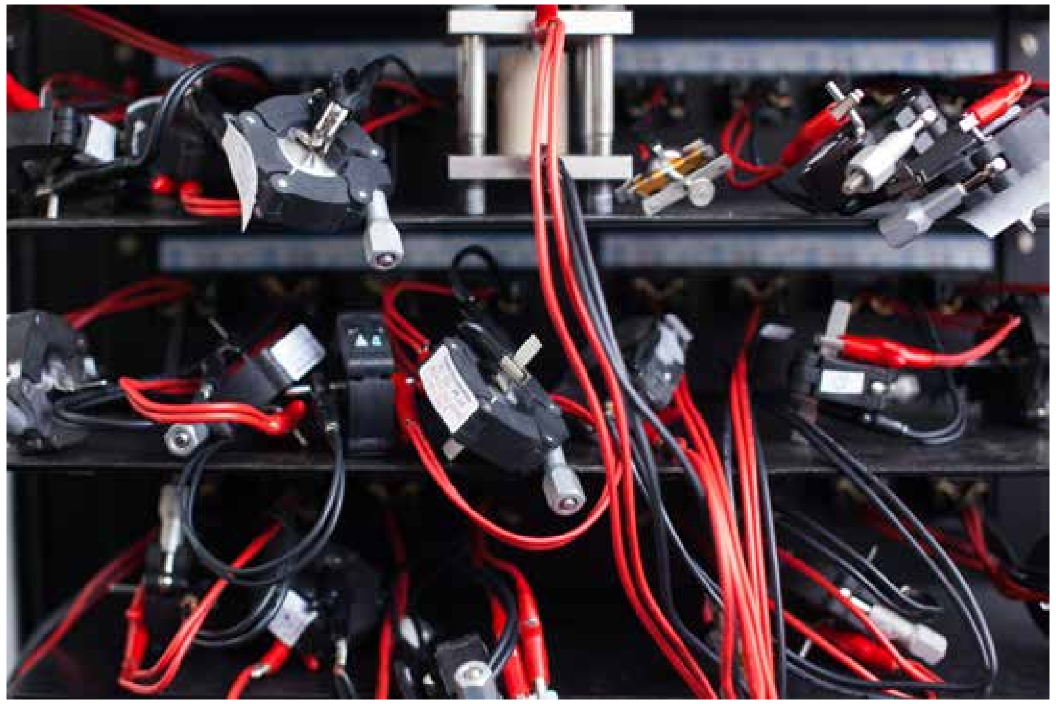


Photo: battery testing facilities in the lab.

Prospects 2020

During the reactor shut down, the "matroesjka" design is being optimized, such that it can be operated in parallel to the classical NDP setup. Additionally, we are in the process of adapting the design of the entire NDP setup such that it can be shifted. This is necessary because of the higher background radiation in the current position. During commissioning, focus will be on the impact of the longer wavelength of the neutrons, which increases the cross section for neutron capture reactions and thus should lead to a higher sensitivity. We believe this will further boost the operando research towards next-generation Li batteries.

Cultural heritage and archaeology

First Imaging Station Holland - FISH

Using a neutron beam as a source for radiography and tomography provides strikingly different measurement capabilities compared to the more mainstream method of using X-rays. Because neutrons interact with atoms in a different way than X-rays, the generated picture has a unique contrast or sensitivity. Some light elements like hydrogen and lithium are extremely well visible with neutrons. Some heavy elements like metals are relatively transparent and the contrast even depends on isotope composition.

The instrument FISH, located in the RID reactor hall, is dedicated to neutron imaging and routinely performs radiography and tomography with a resolution of 100 micrometre. As it is a non-invasive and non-destructive method, it is ideally suited for material studies of cultural heritage.

Progress 2019

In 2019 we continued a project for the Netherlands Institute for Conservation, Art and Science (NICAS). Detailed information about ancient old metal statues like internal composition, casting processes or hidden objects can be revealed with neutron tomography. The picture shows an example where conservation techniques for Egyptian faience were investigated. Other more engineering-oriented applications were in the field of degradation of iron reinforcement in concrete and the investigation of glue thickness between metal plates.

With a good team, a new low-noise camera and control software developed in-house, the instrument has proven to be efficient and reliable. At the same time, new methods developed in the field have been tested and are currently under evaluation for implementation.

After the reactor stop in May 2019, the instrument was completely disassembled and a new detector mounting was developed that should provide better shielding towards the camera. A sample positioning system was implemented, making it easier to scan tall objects like swords or statues in one go.

Prospects 2020

In the summer of 2020, a few months before the start-up of the reactor, we will be able to reassemble FISH again and create a better version than before.

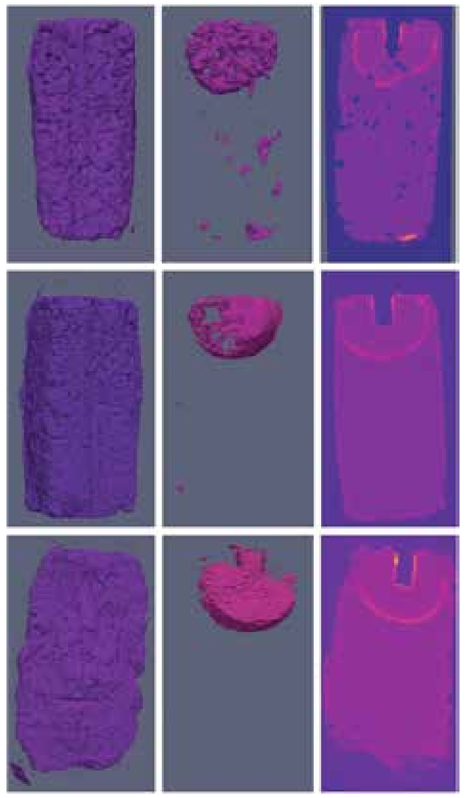


Image:
In the consolidation of Ancient Egyptian faience three glazing techniques were tested to improve mechanical stability. In the example below the penetration depth and distribution of the conservation resin PARALOID B-72 applied to representative faience replicas was investigated. After consolidation, the distribution of the PARALOID B-72 in the replicas was assessed using three-dimensional images created using neutron tomography. Penetration depth was considerably greater in application- and cementation-glazed replicas than in efflorescence-glazed replicas.

Food materials

Spin-Echo Small-Angle Neutron Scattering - SESANS

Spin-Echo Small-Angle Neutron Scattering (SESANS) is a technique that allows users to investigate structures on length scales from 20 nanometres up to about 20 micrometres, which is two orders of magnitude larger than Small-Angle Neutron Scattering (SANS). These length scales are especially relevant for many food materials such as yoghurt and cream cheese. In contrast to traditional scattering methods, SESANS data are in real space, rather than reciprocal space, which makes interpretation easier. These properties make SESANS a powerful tool for the study of the structural properties of materials.

Progress 2019

We studied the effect of enzyme treatments on the wetting behaviour of cotton, as well as the impact of humidity on cotton fibre wall thickness. Supramolecular structures of polymerosomes in different solvents were characterised.

Prospects 2020

We will move the SESANS instrument from the experimental hall to the reactor hall towards the end of the OYSTER programme. We plan to have a solid independent mounting of the foils directly on the table instead of in the magnets. This will give a better control of the orientation of the foils, which is crucial for the magnetic fine-tuning of SESANS. We aim for a better stability of the setup with these changes. We will install an RF flipper in front of SESANS to have a perfect flip of the neutron spin (in contrast to the present V coils that are not ideal).

Photo: the SESANS instrument

Catalysis and magnetocalorics

Mössbauer spectroscopy

Mössbauer spectroscopy is a unique high-resolution technique that has proven to be very valuable for in-situ/operando studies of materials. The high penetrating power of gamma rays makes Mössbauer spectroscopy a very versatile technique to study materials in their working state, revealing the process of catalysis and opening ways to synthesize novel or improved compounds. At the moment, our group in Delft is the only group in the world with the expertise and infrastructure to perform in-situ Mössbauer emission characterization studies under realistic industrial conditions. The characterization of materials and their modifications in the course of their utilization is interesting for many branches of the basic and applied sciences, including physics, chemistry, mineralogy, geology, agronomy and engineering.

Progress 2019

In 2019, the operando characterization of Co/TiO_2 Fisher-Tropsch catalysts under high-humidity conditions was successfully implemented in a PhD project in collaboration with Shell and Eindhoven University of Technology. For our PhD project in collaboration with Johnson Matthey, a high-pressure steam delivery system was developed for the study of water-gas shift catalysts. In a PhD project in collaboration with Wetsus and Kemira, innovative phosphate adsorbents with high adsorption capacity, selectivity and reusability were already identified.

Also, an NWO postdoctoral project was completed in which a neutron in-beam Mössbauer facility concept was developed, increasing the number of usable Mössbauer nuclei. The facility can be used on line. During reactor shutdown, activated Mössbauer sources will also be utilized. Currently, a comprehensive report on the instrument design – exploring concepts such as gamma background, focusing neutron guides and suitable detectors – is being finalized.

Prospects 2020

In 2020 we will start implementing 151-Eu Mössbauer spectroscopy for the characterization of luminescent materials. 121-Sb and 193-Ir Mössbauer experiments on catalytic materials will also be performed in collaboration with Johnson Matthey.

Photo: PhD students Maxim Ariëns and Luke van Koppen preparing in-situ Mossbauer experiments in collaboration with industry partners.

Medical isotopes

OYSTER

Irradiation facilities

Abbreviated as FlexBeFa, the irradiation facilities at the RID are flexible, modular chambers that can be fitted with various types of shielding, depending on which type of radiation is required and which type must be blocked. The facilities are primarily used to produce radioisotopes that find applications in fields such as medicine.

Progress 2019

The flexible irradiation facility has proven to be a successful tool for research purposes as well as for irradiation of holmium microspheres for the treatment of patients with liver cancer. As a result, it was decided to start designing and constructing two pneumatic irradiation facilities. The reason for making the facilities pneumatic is that this allows for transport of samples directly to the lab, thereby avoiding unnecessary radiation exposure of the reactor operators.

Prospects 2020

We will continue realising our plans for the two pneumatic facilities in the research reactor after the CNS works have finished and the research reactor is in use again.

For more information about the use of the flexible irradiation facility in Delft, please read this interview with Jan Sigger, CEO of Quirem Medical. This company uses the facility to produce radioactive microspheres for the radiation treatment of cancer patients.

Photo: Head of the research project Antonia Denkova in one of the labs at the RID.

Faster molybdenum-99 production routes for medical treatment and diagnostics

Radioactive isotopes are widely used in medical diagnostics. For example, about 30-40 million clinical radio-diagnostic scans use technetium-99m worldwide on a yearly basis.

The Department of Radiation Science & Technology investigates the feasibility of producing molybdenum-99 and other useful isotopes by irradiating a continuous liquid target loop system. Compared to the conventional way of irradiating solid targets, this innovative approach supports a continuous and online way of producing, extracting and purifying medical radioisotopes, which is believed to reduce the post-processing time. For example, recent research has shown that the uranyl nitrate solution inside a small, natural circulation driven loop can produce molybdenum-99 continuously for more than 20 years without refilling.

For the purpose of online extraction and purification, innovative microfluidic devices are being explored to selectively extract radioisotopes from the loop. Current research focuses on optimization of the microfluidic system to achieve the maximum extraction and purification efficiency. Recently, a new project entitled “Fast and Efficient Purification of Medical Isotopes by Microfluidic Extraction” has been started in close cooperation with NRG, TRIUMF, URENCO and UMC Groningen. In this project, liquid targets in concurrence with microfluidic devices are being studied under irradiation by our neutrons in Delft and other irradiation facilities.

INTERVIEW

Jan Sigger

Smarter radiation therapy for liver cancer patients

Treatment with tiny radioactive spheres (microspheres) injected directly into the bloodstream can prolong the lives of liver cancer patients who have exhausted other treatment options. Quirem Medical, a spin-off company of University Medical Center Utrecht, developed microscopic spheres that facilitate more accurate imaging support in the treatment of patients. Research conducted at TU Delft has led to improvements in the irradiation of the spheres in the FlexBeFa facility at the Reactor Institute Delft (RID), as a result of which they contain more radioactivity. The RID is now activating these spheres for patient use, while research into second and third-generation microspheres continues.

Race against the clock

Quirem Medical's microspheres consist of holmium-166 packed in a kind of biodegradable plastic. Jan Sigger, CEO of Quirem Medical: "The radioactivity of the microspheres naturally decreases after irradiation due to the half-life of holmium-166, so getting them to the patient with enough activity is a race against the clock. What makes the flexible irradiation facility (FlexBeFa) in Delft unique is that we are able to irradiate the spheres for much longer, thereby achieving a high level of activity. This gives us more time to process and transport the radioactive microspheres. In their research programme, head of the research project Antonia Denkova and her colleagues have developed an irradiation facility in which various types of shielding can be applied. This allows the holmium to be irradiated for longer periods of time without the outside of the spheres being damaged. While we were hopeful when we started this research, the results have exceeded our expectations. It gives much more insight into the optimal neutron activation." In the FlexBeFa facility, as well as producing holmium-166, Denkova's team is also researching possibilities for other radioisotopes.

Opening the black box

The technique of radioembolisation is not new, but Quirem Medical's holmium-166 variation has a unique advantage. Sigger: "As well as a therapeutic beta radiation, the holmium in our microspheres emits gamma radiation and is also paramagnetic. Doctors can therefore accurately map the distribution

"We will be able to improve prospects worldwide for liver cancer patients who have exhausted their treatment options, and hopefully also contribute to the treatment of patients with other tumours in the future."

of the microspheres using both SPECT (nuclear imaging) and MRI. So you can see precisely where the spheres are after injecting them. You could compare it to dropping a leaf in a river: you don't know exactly which river branch it will end up in. With radioembolisation, you hope that the radioactive spheres find their way into the capillaries of the tumour via the blood so that the radiation destroys the cancer cells there. In the past, a patient was sent home and it was not known until a few weeks later whether the treatment had any effect. Physicians more or less treated their patients in a black box. Now we can see the location of the spheres immediately."

New research in Nijmegen

Radboudumc in Nijmegen has recently started a new study, led by Frank Nijsen in collaboration with Quirem Medical, into real-time imaging of spheres. "The first treatments were successful and MR imaging during administration went well. By observing during treatment, we learn a lot about how the spheres distribute. This makes it possible to very accurately inject the right dose around and into the liver tumours. The research in Nijmegen will therefore also have an effect on the research in Delft. The amount of radiation activated in Delft can then be varied from one patient to another. Ultimately, the idea is to insert the spheres very accurately into the tumour itself, thus sparing as much healthy liver tissue as possible. If a tumour needs more radioactivity, we can administer more spheres. This adaptive approach will be the next important step in the research."

Great medical need

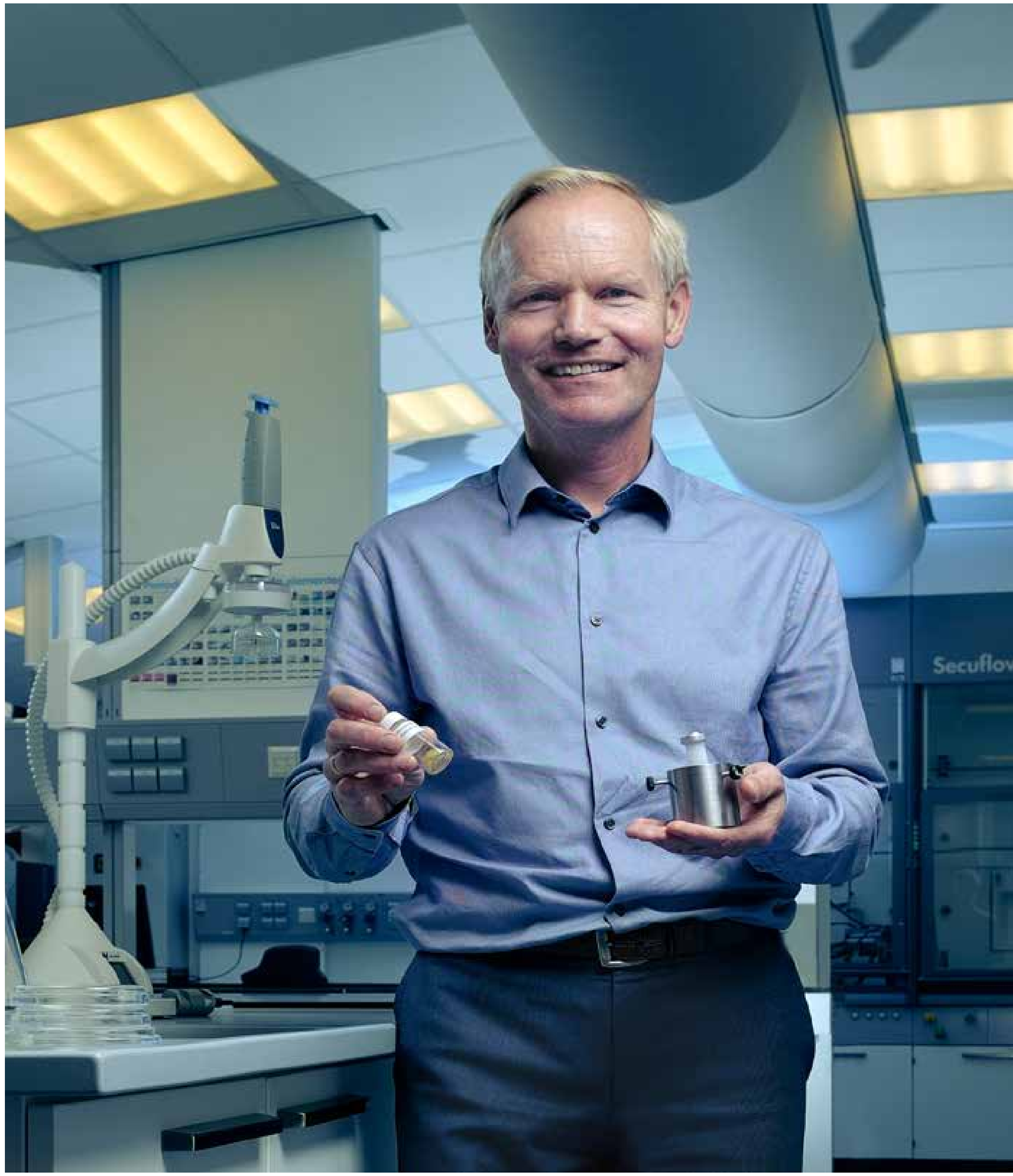
Meanwhile, researchers from Quirem Medical, Radboudumc and the RID are already working on the second and third-generation microspheres. "There is a great medical need, also for brain tumours and pancreatic cancer, for example," Sigger says. "We currently administer the spheres through the bloodstream, but we are also exploring the possibilities of injecting the microspheres directly into the tumour. In addition, we are looking at increasing the holmium content, which would allow us to deliver a higher dose

"Thanks to the unique conditions of the Delft research reactor, it will be possible to load the spheres with more radioactivity, enabling us to supply them to Asia and the United States. "

with fewer microspheres. While it is still preclinical research at the moment, I am hopeful that in five years' time we will have the first positive clinical results for the use of our microspheres in, for example, pancreatic tumours and brain tumours."

Unique conditions in Delft

One of the two new FlexBeFa facilities being built in Delft will be used largely for the activation of the holmium microspheres. Sigger: "An inherent part of working with reactors is that they are occasionally shut down for maintenance. The reactor shutdown in Delft is taking longer due to the OYSTER programme. I am looking forward to the reactor being available for patient treatment again. Thanks to the unique conditions of the Delft research reactor, it will be possible to load the spheres with more radioactivity, enabling us to supply them to Asia and the United States, for example, whereas we are currently active mainly in Europe and Turkey. The research and development climate in the Netherlands is very favourable for us. We work closely together with the RID and use a hot lab for processing radioactive products at the Sint Franciscus Gasthuis hospital in Rotterdam, just a 15-minutes' drive from the RID. The government is also willing to invest in our innovations with grants and funds. As a result, we will be able to improve prospects worldwide for liver cancer patients who have exhausted their treatment options, and hopefully also contribute to the treatment of patients with other tumours in the future."



CEO Quirem Medical: Jan Sigger.

Photo credits: Marc Ilford fotografie

International developments: the European Spallation Source (ESS)

The European Spallation Source (ESS) is a European Research Infrastructure Consortium (ERIC), a multi-disciplinary research facility based on what will be the world's most powerful neutron source. Since 2014, the ESS is under construction on the outskirts of Lund, a city in southern Sweden. The facility's unique capabilities will both greatly exceed and complement those of today's leading neutron sources, enabling new opportunities for researchers across the spectrum of scientific discovery, including materials and life sciences, energy, environmental technology, cultural heritage and fundamental physics. Many countries within Europe have already partnered up with the host countries Sweden and Denmark. The Netherlands, one of the early supporters of ESS, has not made a formal financial commitment yet and holds its official status of Observer pending the completion of the required national procedures within the Netherlands.

Photo: The ESS-construction site in Lund, Sweden.

Integrated Management System

The Integrated Management System (IMS) for RID forms a coherent system of processes, procedures, work instructions and performance indicators indicating the contributions of each component of the RID organisation. Based on the 'plan-do-check-act' (PDCA) cycle, the IMS ensures that the safety, legal, licence and customer requirements are met. For RID in general and for OYSTER specifically, these are important matters.

Progress 2019

In 2019, a meeting was held with the Dutch Authority for Nuclear Safety and Radiation Protection (ANVS) to provide insight into the current state of the implementation of the IMS. The conclusion of this meeting was that the design and review of the current work processes have been carefully carried out. Partly through workshops and brown-paper sessions, RID staff learned more about the IMS. The work processes form the core for the design and implementation of the IMS. The work process relating to the preparation of work with third parties was drawn up.

Extra attention was paid to stimulating the use of the document management system by means of information and communication. The website is easy to find and is being used more and more often. The house rules have been reviewed and accessibility and legibility were improved.

The internal audit process was evaluated and the audit schedule properly carried out.

Prospects 2020

In 2020, new internal auditors will be trained so that we can continue carrying out the audit schedule properly. In order to ensure proper operation and maintenance of the CNS installations, instructions and procedures will be drawn up for this purpose.

Financial overview

OYSTER

The available OYSTER project budget amounts to €117 million, covering the initial investments as well as the basic reactor-associated operational costs for a period of 10 years. In 2012, the Dutch government awarded €38 million for OYSTER. TU Delft contributes a total of €74 million in kind.

Furthermore, TU Delft stands surety for an additional €5 million. This is part of the co-funding (industrial, scientific etc.) needed to fund the development, commissioning and exploitation of instruments and facilities over the total 10-year OYSTER programme period and beyond.

Update 2019

The work of various subcontractors was delayed in 2019. This was mainly due to bankruptcy of one supplying party, and deliveries that did not meet the specifications. As a result of the delays, it was decided to shift the official end date of the project to 30 September 2020. The delays caused by the selection of new subcontractors and the failure to meet the specifications entail an additional €1.75 million. This overrun is caused by extending the contracts of extra personnel and the additional costs incurred by the change of suppliers. Because these costs can no longer be covered within the current budget, TU Delft has had to make a provision for this amount.

To properly monitor the progress of the project, a risk matrix has been drawn up which is discussed weekly in the core team meeting of OYSTER. In addition, an overall project planning has been drawn up in which all disciplines are included for the period up to and including the commissioning of the instruments. This will ensure that the critical path is clear to everyone involved.

In total, €25 million was spent on the project until the end of 2019: €19 million for OYSTER and €6 million related to the new safety requirements legislation (DSR). The in-kind contribution from TU Delft was €11 million in 2019, including the additional provision of €1.75 million

Instrument developments

Overall, the main priority in 2019 was given to the instruments that ‘view’ the cold source. The Small-Angle Neutron Scattering (SANS) instrument underwent significant detector maintenance programme and the neutron reflectometer (ROG) is undergoing a major upgrade to prepare it for optimal use. The Neutron Depth Profile (NDP) instrument is also being prepared for cold neutron operation with the ordering of a new neutron guide and the design of a new position. The Spin-Echo SANS (SESANS) instrument is being prepared for movement to another beamline to make room for cold neutron imaging that is under design as well. Connected to the operation of these instruments, two persons were hired on instrument scientist/assistant professor (tenure track) positions. They will be responsible for the daily running of the SANS and ROG instruments. For the irradiation facilities, we have officially started to create a second flexible irradiation facility (FlexBeFa) with a pneumatic station.

Planning

OYSTER

The schedule for executing the OYSTER project changed. The project end date has been extended until the end of September 2020 mainly due to delays in the works of subcontractors. One of the reasons for the delay is a change in one of the subcontractors and as a result of the bankruptcy of another.

Task name	Contractor	2019 Q1 J	F	M	Q2 A	M	J	Q3 J	A	S	Q4 O	N	D
OYSTER Project		■	■	■	■	■	■	■	■	■	■	■	■
Milestones													
KEW Nuclear License					■								
Final Performance Test													
Project Finish													
CNS Utilities													
Civil & Infrastructure (Bridge)	Strukton												
Electrical Systems	Worksphere												
System Descriptions	KAERI - RID	■	■	■									
Helium Refrigeration System	Stirling Cryogenics	■	■	■									
Helium Transfer Lines	Demaco												
Vessels & Tanks	Kreber	■	■	■	■	■	■	■	■	■	■	■	■
Gas Blanket Systems	PFS - Demaco	■	■	■	■						■	■	■
Instrumentation & Control	Yokogawa	■	■	■	■	■	■	■	■	■	■		
Reactor Modifications													
Test In-Pool Assembly (Mock-up)	Moojin Keeyeon	■	■	■	■	■	■	■	■	■	■	■	■
Main In-Pool Assembly	Moojin Keeyeon	■	■	■	■	■	■	■	■	■	■	■	■
Beam Tube Modification	Bilfinger - Babcock Noel	■	■	■	■	■	■	■	■	■	■	■	■
Reactor Protection System	Yokogawa	■	■	■	■	■	■	■	■				
Primary & Secondary Cooling System	RHDHV / Stork	■	■	■	■	■	■	■	■	■	■	■	■
Commissioning													
Procedures & Protocols	KAERI										■	■	■
System Performace Test (SPT)	KAERI - RID												
Integrated System Test (IST)	KAERI - RID												
Reactor Integrated Test (RIT)	KAERI - RID												
Training & Documentation	KAERI - RID							■	■	■	■	■	■

Task name	2020 Q1 J	F	M	Q2 A	M	J	Q3 J	A	S	Q4 O	N	D
OYSTER Project	■	■	■	■	■	■	■	■	■	■	■	■
Milestones												
KEW Nuclear License												
Final Performance Test									■			
Project Finish									■			
CNS Utilities												
Civil & Infrastructure (Bridge)		■	■									
Electrical Systems				■	■	■						
System Descriptions												
Helium Refrigeration System												
Helium Transfer Lines				■	■	■						
Vessels & Tanks	■	■	■									
Gas Blanket Systems	■	■	■	■	■	■						
Instrumentation & Control			■									
Reactor Modifications												
Test In-Pool Assembly (Mock-up)	■	■										
Main In-Pool Assembly	■	■	■	■	■	■						
Beam Tube Modification	■	■	■									
Reactor Protection System												
Primary & Secondary Cooling System	■	■	■	■	■	■	■	■				
Commissioning												
Procedures & Protocols	■	■	■	■	■	■	■	■				
System Performace Test (SPT)				■	■	■	■					
Integrated System Test (IST)								■				
Reactor Integrated Test (RIT)									■			
Training & Documentation	■	■	■	■	■	■	■	■	■