03 Technology geared towards innovation

First of a kind for BR2

Fusion requires high-neutron-flux irradiation at 1200˚C

Two decades into the future seems a long way off. But at Cadarache in the South of France, the test fusion reactor ITER is currently being built. This is the last step before the construction of the industrial fusion reactor DEMO, which should be ready by about 2044. As a research centre, SCK•CEN is also doing its bit: for the European programme EUROfusion, high-flux irradiation will take place in BR2 at 1200°C. Such extreme conditions have never before been achieved in the history of SCK•CEN.

The ITER reactor, which will be arriving at Cadarache in France within a few decades, is the first step towards demonstrating both the technical and the commercial benefits of nuclear fusion. Research teams are currently developing and specifying the materials that will be contained in the fusion reactor, including those for the 'first wall', which will be directly exposed to the plasma.

The selection of basic materials for use in ITER has now been completed. Tungsten will be used as the armour material for the divertor – a component that maintains a maximum heat load in order to achieve the most stable discharge of the plasma. The end-of-life dose received by the divertor material in ITER will not exceed 1 dpa (displacement per atom).

Research into new materials now needs to be developed to ensure that commercial fusion facilities can be operated safely – specifically DEMO, the industrial fusion prototype that will be the successor to ITER. The accumulated dose will be at least a factor of 10 or 20 higher. This nuclear fusion research is being carried out as part of the EUROfusion project, a H2020 partnership between Euratom and a consortium of various European Union member states, Switzerland and Ukraine.

"The first irradiation campaign above 1000°C in BR2 will be a real challenge and will provide new opportunities for the development of high-temperature materials for both nuclear fusion and nuclear fission."

Irradiation campaign in BR2

At SCK•CEN, the initial data is being screened and selected for irradiation conditions 'after ITER'. In 2017, the researchers are due to start a new irradiation campaign in the research reactor BR2 in order to select innovative tungsten-based materials. It is possible to mimic fusion conditions in BR2 thanks to its core flexibility, the high flux and the wealth of in-house expertise available. By increasing the neutron flux and the irradiation temperature, the researchers create neutrons that are similar to those in a fusion environment. However, the end-of-life conditions require a long uninterrupted period of irradiation of at least five years.

New irradiation equipment

The project will run for about three years and has a total budget of 2.5 million euros, half of which will be used for irradiation, and one-third for post-irradiation experiments (PIE). The project also includes the development of new irradiation equipment, the High Temperature High Flux (HTHF) (see box).

In this machine, high-neutron-flux irradiation will be carried out at 1200°C under active temperature and environmental control, a first in the history of experiments conducted at SCK•CEN. After irradiation, the researchers will systematically document the thermal, mechanical and micromechanical properties of the irradiated materials in SCK•CEN's Laboratory for High and Medium Activity (LHMA).

The materials to be irradiated are tungsten-based alloys, designed for the first wall and the armour. The researchers use nano-engineering to achieve the best results under fusion conditions. Their aim with the irradiation campaigns is to investigate whether the performance of the production materials will be maintained after exposure to neutrons or whether these neutrons will throw a spanner in the works.

> *"The project also comprises the development of a new irradiation device, the so-called High Temperature High Flux (HTHF)."*

Step by step

The project itself consists of three parts with various post-irradiation tests and is proceeding in collaboration with the research centres FZJ and KIT (Germany) and Demokritos (Greece). There is good reason to share the project among different centres. One single laboratory does not have all the requisite materials, expertise and knowledge in-house. Furthermore, it is a race against time, because the schedule for establishing the DEMO design is really tight, even though the plant is not due to be completed until 2044.

GRAPHITE KEEPS IT COOL

HTHF is the name of the housing in which the TUNER project (TUngsten NEutron irRadiation) will carry out specific irradiation. This specific housing will be used to investigate the effect of the combination of high temperature (800°C) and high flux (1 dpa in tungsten) on materials and their properties.

The advantages of graphite

Graphite was chosen because it has a low neutron-absorption capacity and good thermal conduction properties. Furthermore, due to its low specific gravity, graphite warms up less of its own accord. In this way, the researchers make sure that the temperature does not increase too much. Initially, the design followed a few more general criteria; at a later stage, it was tailored to the client's specific criteria. Designer Gitte Borghmans had to work in an extremely detailed manner: 'There are 26 cases, hanging one after the other like small carriages. Each case is only 30 mm high.'

Six cycles in BR2

The HTHF equipment has now been designed and construction will go ahead in early 2017, so that irradiation can commence before the 2017 summer holidays. As a result of the required 'high flux', HTHF will remain in the BR2 reactor over six cycles, which is approximately a year. The LHMA laboratory will analyse the irradiated samples in 2018.

The BR2 campaign is going to be a real challenge and will reveal whether BR2 is able to provide extreme irradiation conditions for new materials. After successfully demonstrating the HTHF performance, the next stage in the research consists of a sub-miniaturisation programme: the smaller the volumes, the lower or cheaper the irradiation conditions that need to be met. Small sample volumes also mean quick deactivation, inexpensive transport and, generally, fast PIE feedback. In the future, the use of miniaturisation will grow significantly, which will be the next big challenge for the Fusion Materials Programme.

Technology

Sustainability is not a buzzword

Thanks to its refurbishment and unique set-up, BR2 is still one of the best test reactors and will continue to be for generations to come. BR2 is also a very reliable source of medical radioisotopes, accounting for as much as two-thirds of global production. The further development of new and promising medical radioisotopes for cancer treatment shows that our nuclear research is and will always be characterised by sustainability.

Leo Sannen
Nuclear Materials Sciences Institute Director

How does heat affect pore water composition in the Boom Clay?

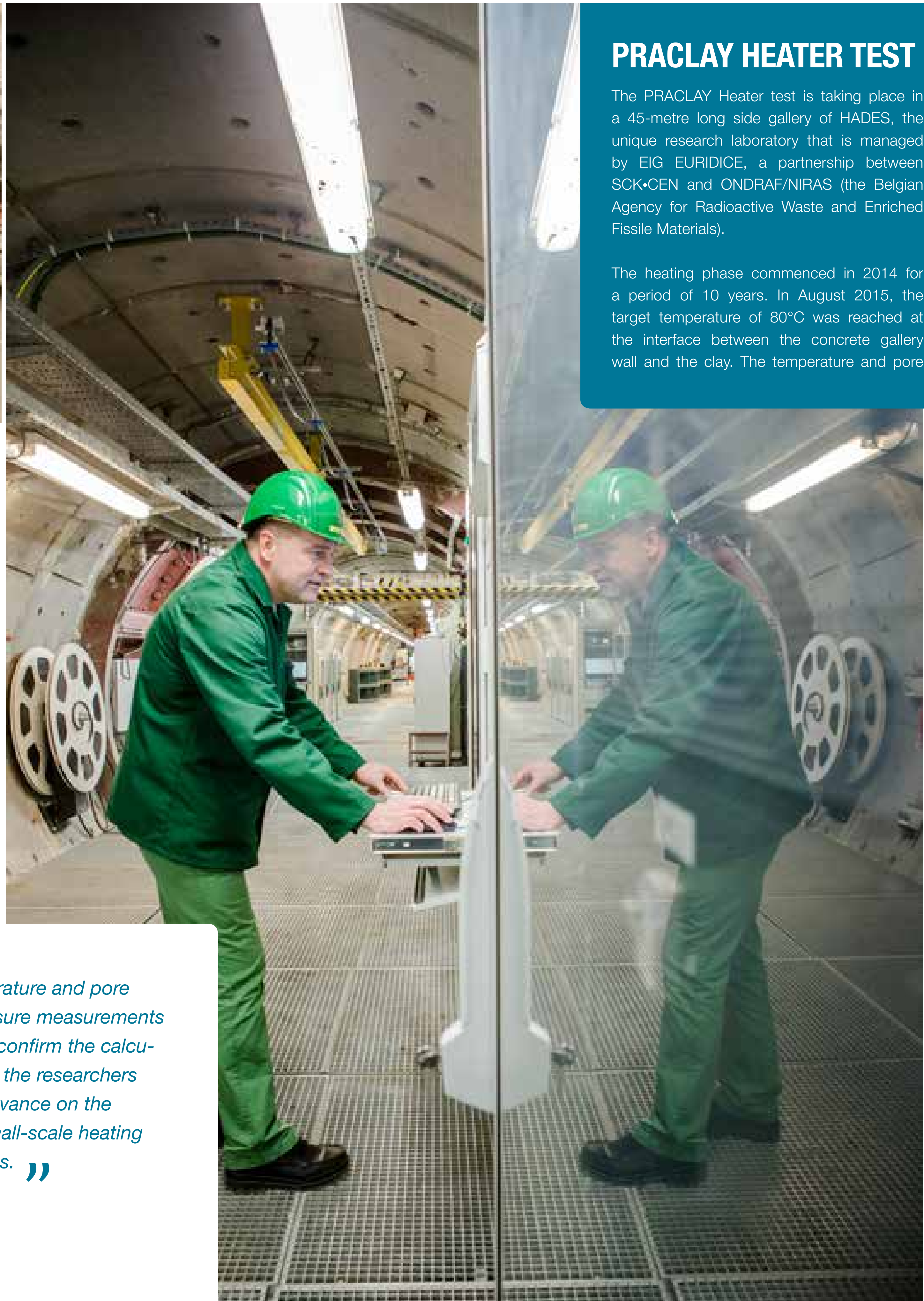
Research 225 metres underground

Is the Boom Clay suitable for underground disposal of radioactive waste? This question is currently the subject of intensive research. One project is investigating the extent to which the heat emitted by radioactive waste – simulated by the PRACLAY Heater test – influences the composition of pore water, which can, in turn, influence the behaviour of radionuclides. The experiments are being carried out in the HADES laboratory, 225 metres below ground level.

Scientists around the world consider deep underground disposal in geologically stable strata to be the most appropriate way to manage highly radioactive waste in the long term. In Belgium, the research programme is focusing on deep clay formations such as the Boom Clay. Before disposal in clay can become a practical reality, however, the behaviour of the clay and the impact of possible disruptions must be thoroughly investigated. SCK•CEN started this research in Belgium almost 40 years ago.

Research into the impact of heat on the composition of pore water takes place in the underground HADES laboratory, as part of the PRACLAY Heater test. This test enables researchers to investigate how the Boom Clay behaves thermally, hydromechanically and chemically when it is heated as a result of contact with highly radioactive waste.

Researchers **Mieke De Craen** and **Miroslav Honty** in the underground laboratory HADES



Research into thermal load

The aim of one of the experimental set-ups is to study the chemical changes that occur as a result of the clay warming up. The research will continue over a period of 10 years. What does it actually involve?

Researchers Mieke De Craen and Miroslav Honty take us 225 metres deep underground: 'We analyse the pore water and the gases dissolved in it at regular intervals in the laboratory. To do this, we drill a hole in the clay and insert a piezometer. This is a type of metal tube with filters that collect the water at various points. That water is conveyed to the experimental set-up in the underground laboratory via thin pipes. Under normal circumstances, the pore water at this depth has a temperature of 16°C, but as a result of the heating phase of the PRACLAY Heater test, the temperature at the filters is currently around 55°C. This can disturb the chemical equilibrium and that's exactly what we want to study. The experimental set-up enables us to measure a number of specific parameters and to sample the pore water and the gases dissolved in it separately. The samples are then sent to the above-ground laboratory for further analysis.'

> *"The temperature and pore water pressure measurements in the clay confirm the calculations that the researchers made in advance on the basis of small-scale heating experiments."*

PRACLAY HEATER TEST

The PRACLAY Heater test is taking place in a 45-metre long side gallery of HADES, the unique research laboratory that is managed by EIG EURIDICE, a partnership between SCK•CEN and ONDRAF/NIRAS (the Belgian Agency for Radioactive Waste and Enriched Fissile Materials).

The heating phase commenced in 2014 for a period of 10 years. In August 2015, the target temperature of 80°C was reached at the interface between the concrete gallery wall and the clay. The temperature and pore water pressure measurements that have so far been carried out in the clay confirm the calculations made by the researchers in advance on the basis of the results of earlier small-scale heating experiments. This confirmation, on a scale that is representative of an actual waste repository, is one of the most important aims of the experiment. In addition, the PRACLAY Heater test also provides the opportunity to assess the geochemical changes in the clay, the stability of the concrete lining and the reliability of the measuring instruments at elevated temperature.

Comparing results with predictions

Heating causes chemical reactions that affect the pore water. The researchers want to determine the extent to which this happens and whether it has an impact on the behaviour of radionuclides. Mieke De Craen and Miroslav Honty again: 'We have a large database of pore water data at a naturally occurring temperature of 16°C. And we also have geochemical models for predicting the composition of pore water at increased temperatures. But until now, we haven't had the opportunity to test their validity using experimental data.'

This is now possible on a large scale thanks to the HADES heating experiment. The researchers will determine the pore water composition and compare the results with their predictions: 'Thanks to the new data, we'll be able to study the situation in detail at elevated temperatures. A study is also currently under way in close collaboration with the Microbiology group. They are going to assess the possible presence and activity of micro-organisms in pore water in the Boom Clay subject to thermal stress.'

U_3O_7: the missing piece of the puzzle

A better understanding of the uranium dioxide oxidation process

Uranium dioxide (UO_2) is the most commonly used nuclear fuel. When exposed to air or water, UO_2 oxidises to U_3O_8. During this oxidation reaction, the material swells by at least a third, and with very fine powders this process can take place very quickly. Scientists have spent more than seventy years studying the complex structures that are formed during the oxidation of UO_2 to U_3O_8, but an important intermediate phase, namely U_3O_7, has remained unexplained until now. Gregory Leinders changed all that with his PhD research.

Uranium powders and cylindrical fuel pellets always oxidise if they come into contact with air. This can have drastic consequences for both the production of nuclear fuel and the storage of spent fuel. During the oxidation reaction, a considerable amount of heat is released, as a result of which there is a risk of auto-ignition of very fine powders. This generates a number of safety risks, because fire is to be avoided at all costs in a production process.

And that's not all. When the oxidation reaction goes to completion, the higher and more stable oxide U_3O_8 is formed. This oxide cannot be used for the production of nuclear fuel. During the crystallographic transition to U_3O_8, the volume increases by as much as 36 per cent. This can cause storage containers to crack, during not only the production process, but also the processing and storage of the spent fuel.

Contradictory information

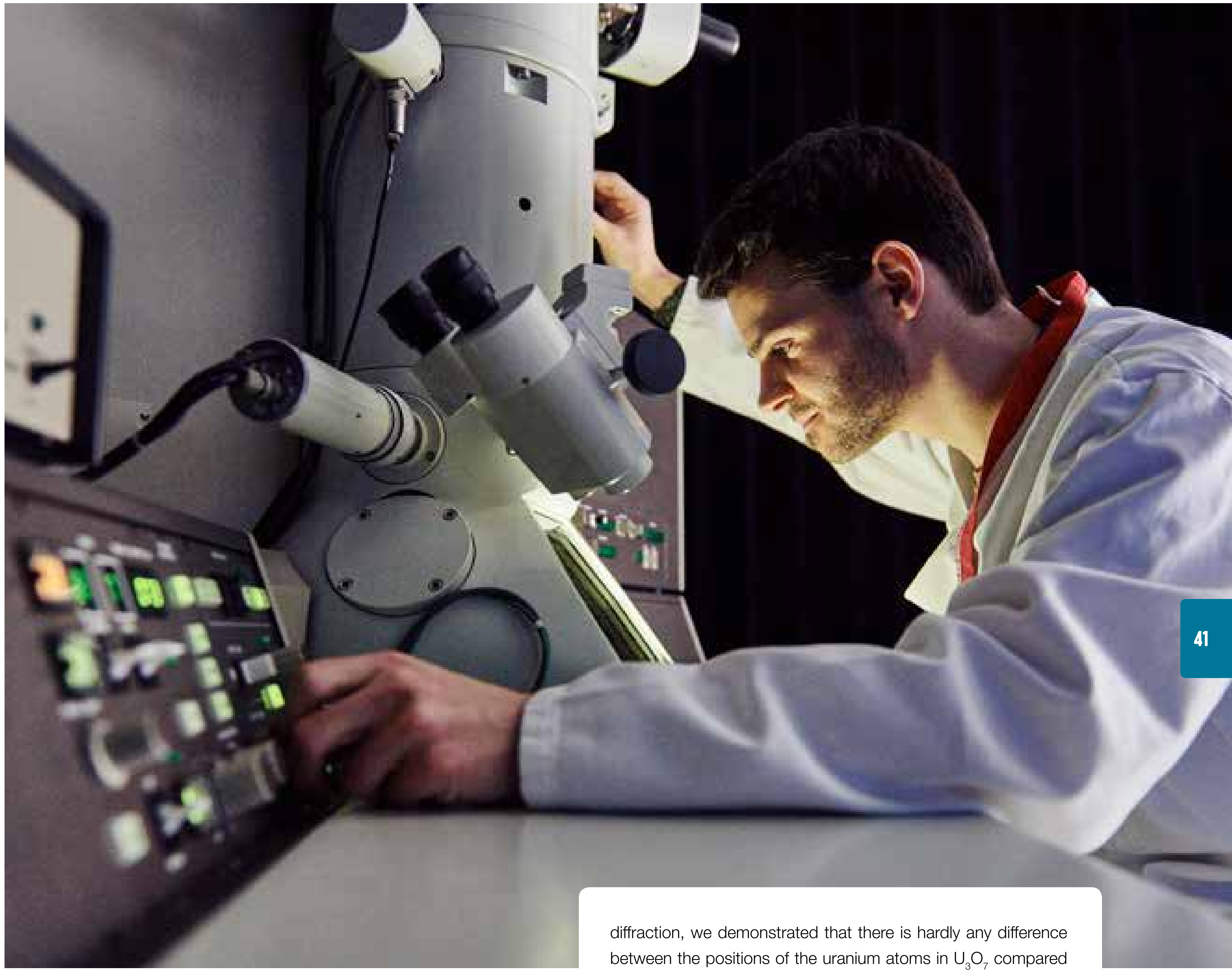
Together with his colleagues from the Fuel Materials group, Gregory Leinders investigated how oxygen interacts with UO_2 powders: 'I looked at the way in which and how quickly oxidation occurs. The conditions were based on those in the production of nuclear fuel and the storage of UO_2 powders and fuel pellets. Specifically, a maximum oxygen concentration equal to that in the air and temperatures of up to around 250°C.'

Before U_3O_8 is formed in these conditions, the intermediate uranium oxide U_3O_7 is created. Gregory Leinders: 'Although we've known about this oxide for seventy years, we didn't understand its complex crystal structure. The data was sometimes even contradictory. My colleagues and I managed to develop a consistent model for this structure, based on new experimental results.'

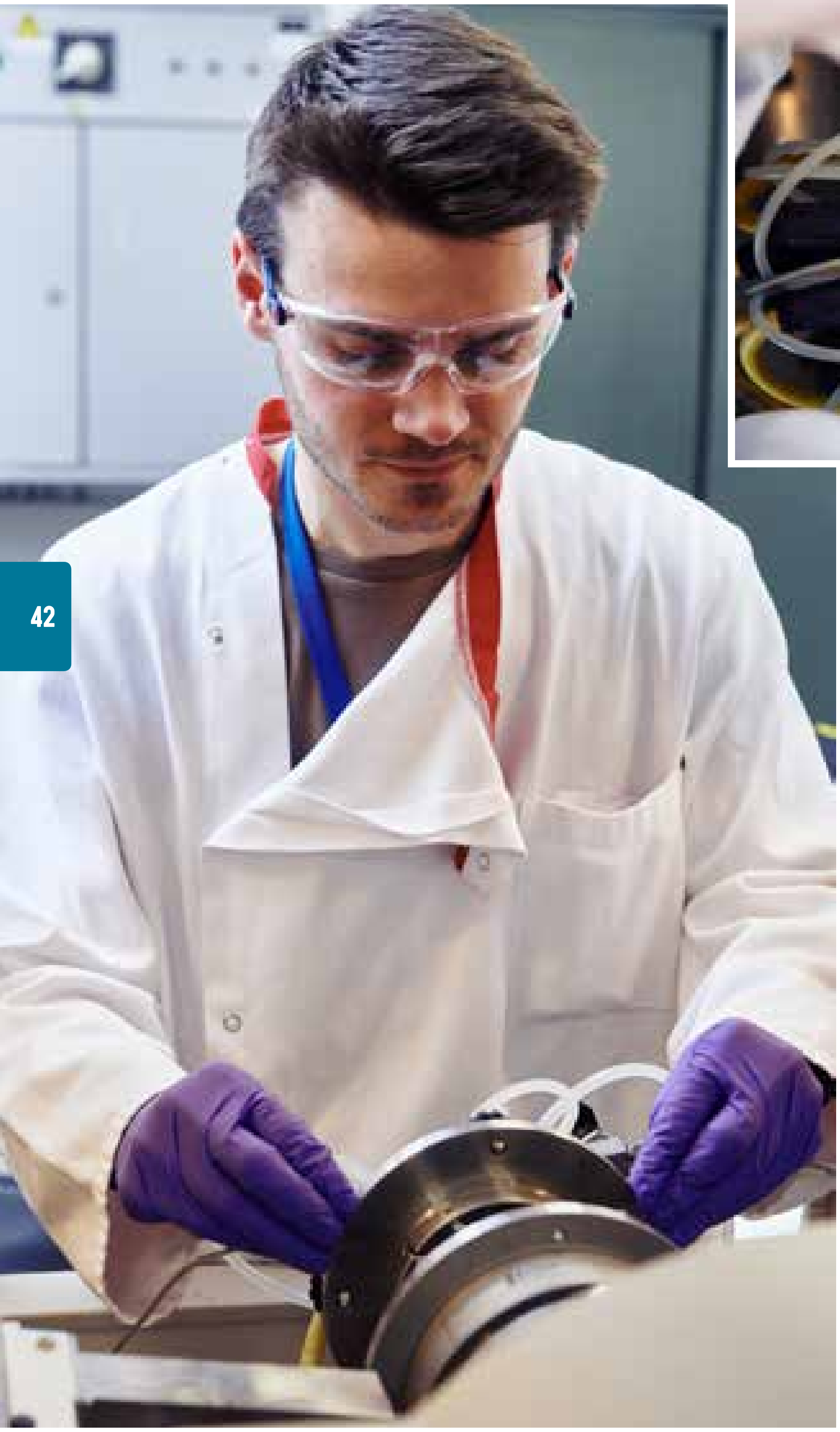
Periodic arrangement

After polycrystalline U_3O_7 powders were produced in the SCK•CEN labs, researchers used X-ray diffraction and electron diffraction for the structural analysis: 'We do that using a beam of X-rays and a beam of electrons, so that a scattering process takes place that is sensitive to the position of the atoms in the structure. The result is an interference pattern, and that's what we measure. Diffractograms like this provide us with information about the position of atoms and the distance between them, and also about the symmetry of the crystal structure.'
The use of the two diffraction techniques made it possible to determine the structure properly: 'Using X-ray diffraction, we demonstrated that there is hardly any difference between the positions of the uranium atoms in U_3O_7 compared with UO_2, while electron diffraction enabled us to determine the clustering of the additional oxygen atoms. On average, the structure closely resembles that of UO_2, but additional anions are grouped into cuboctahedral oxygen clusters, which results in a long periodic arrangement. The crystal lattice can subsequently be described as an enlarged primitive cell that contains 15 fluorite-type subcells.'

Scientists have been racking their brains for over 70 years to solve this problem. SCK•CEN researchers have finally found a consistent model for for the complex crystal structure of U_3O_7.



“We finally developed a consistent model for the complex crystal structure of U_3O_7.”



Towards a quantitative interpretation

Having determined the U_3O_7 crystal structure, SCK•CEN has discovered an essential missing piece of the puzzle of the oxidation mechanism of UO_2. But more work is needed: ‘The primitive cell in itself still requires more detailed research. In a follow-up study, we want to figure out additional structural information using state-of-the-art techniques. These include synchrotron X-ray absorption spectroscopy (XAS), in order to establish the valence state of the uranium atoms. Another option is precession electron diffraction to further refine the crystal structure – this is a more advanced technique that can be used to determine and interpret intensities in an electron diffractogram quantitatively. All of these results should contribute to a greater understanding of the way in which the important end product of oxidation is formed: U_3O_8.’

New opportunities in cancer treatment

Valuable capsules with thorium for Targeted Alpha Therapy

The past can hold valuable treasures for the future. In the 1970s, SCK•CEN produced sources of thorium and is currently one of the rare research institutes in the world to possess thorium-229, a scarce and promising radioisotope. Today, the new SERAPHIM project is picking up where they left off: thorium can make a useful contribution to cancer treatment.

Thorium-229 (Th-229) is a valuable radioisotope, used in atomic clocks, for example. But that’s not all: the daughter isotopes actinium-225 (Ac-225) and bismuth-213 (Bi-213) have a great deal of potential for cancer treatments. That sounds encouraging, but there is one problem: the global quantity of Th-229 is extremely limited. SCK•CEN has succeeded in demonstrating that there are relevant quantities of Th-229 in the historical sources of thorium, creating the opportunity to initiate R&D into radiopharmaceuticals. Researchers are specifically interested in the decay product Ac-225, both for direct applications and for the creation of a generator for Bi-213.

Targeted alpha therapy

Ac-225 and Bi-213 can be coupled to specific antibodies. An antibody is a type of carrier molecule to which a short-lived radioactive particle can be attached. The antibody coupled to Ac-225 or Bi-213 subsequently moves inside the body and binds specifically to a cancer cell like a key in a lock, and during radioactive decay of Ac-225 and Bi-213, alpha particles are released that destroy the cancer cell. This is the principle of targeted alpha therapy (TAT).

One of the few

SCK•CEN is one of the few research institutions in the world that is in possession of Th-229 as a source for Ac-225 and Bi-213. During research into the historical sources of thorium, the available quantity of Th-229 was established using a non-destructive measurement method based on gamma spectrometry and mathematical modelling. The qualitative method applied had the advantage that the hermetic seal on the historical capsules remained intact and that the purity of the valuable product was not compromised. The knowledge and experience of the Dismantling, Decontamination & Waste expert group was crucial when it came to determining the test set-up and interpreting the results. The thorium – just 1 milligram! – was contained inside capsules that needed to be opened with great care in a subsequent phase.

“SCK•CEN is one of the few research institutions in the world that is in possession of Th-229, a precious radioisotope.”

“Through our preclinical research, we want to convince the medical community of the huge benefits of targeted alpha therapy. This insight may open the door to large-scale isotope production for SCK•CEN. It’s very promising!”

Mission accomplished

The capsule was successfully opened in December 2016. Work immediately commenced in the form of the SERAPHIM project (Separation of thorium-229 from historical sources for the production of radioisotopes for targeted alpha immunotherapy). First of all, the radiological content is being processed to separate out Th-229. After that, the aim is to produce an Ac-225/Bi-213 generator, from which Bi-213 will be extracted for the development of Bi-213-coupled antibodies for the treatment of ovarian and breast cancer. This is being carried out as part of the PhD research of Yana Dekempeneer, which involves a successful collaboration between the Radiochemistry expert group and the Vrije Universiteit Brussel.

Thomas Cardinaels, head of the project, firmly believes that these alpha isotopes have a great future ahead: ‘Through our preclinical research, we want to convince the medical community of the huge benefits of targeted alpha therapy. This insight may open the door to large-scale isotope production for SCK•CEN. It’s very promising!’