In the loop : developing the next generation of reactors

In the loop

Professor Martin van Sint Annaland is breaking new ground in the development of novel power producing technologies. Here he talks about his research, explaining why it could prove crucial in reducing anthropogenic climate change

By way of an introduction to your research, can you explain chemical looping combustion (CLC)? What are your key objectives within this field of study?

CLC is a way to carry out the combustion of a fuel (either natural gas, coal, biomass or their derivatives) without the air and fuel coming into direct contact. This is possible by making use of a solid material – the oxygen carrier – which is first oxidised with air, thus harvesting oxygen from the air, and then reduced by a fuel, thereby releasing the oxygen required for combustion. The cycle of oxidation and reduction is then repeated.

During oxidation, the gas stream leaving the reactor is chiefly comprised of nitrogen, whereas during the reduction stage the gas stream mainly consists of CO_2 and water. In this way, unlike in traditional power plants, CO_2 separation is automatically achieved as part of the standard operation of the reactor. Because of this, the cost of electricity production with integrated CO_2 capture can be significantly decreased.

Can you discuss the key advantages of using CLC systems in conventional power stations?

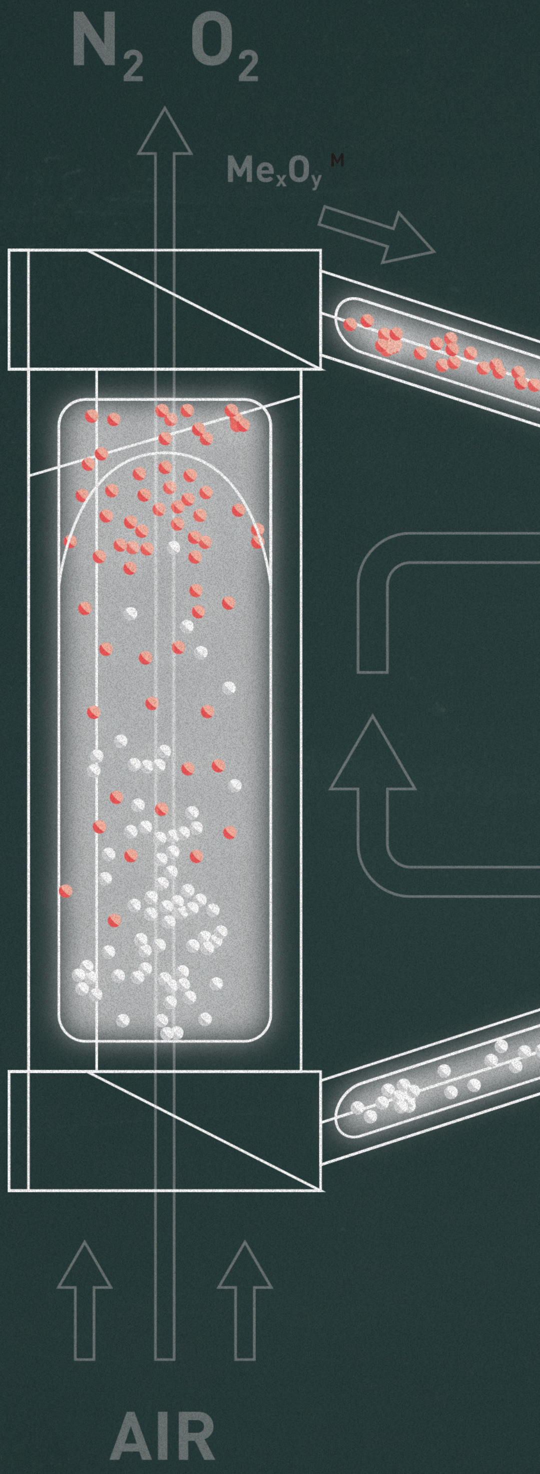
Conventional power stations combust fossil fuels to produce power and emit a gas stream in which nitrogen is mixed with CO_2. Separation of this CO_2 from the flue gas is very costly and energy intensive. On the other hand, with CLC the separation of the CO_2 is inherent to the reactor's operation. So a power plant based on CLC operating at high temperatures and pressures will achieve much higher efficiencies than a conventional plant which uses traditional CO_2 capture. Additionally, the CO_2 produced during CLC is extremely pure, and the amount of CO_2 separated is very high, further increasing the efficiency of the downstream processing of the gas.

What have been the drawbacks of CLC technology, preventing its uptake into power plants? How have you worked to overcome these disadvantages?

Performance of the power plant is only improved using this technology if CLC is operated at high temperatures and pressures, approximately 1,200 °C and 20 bar. For this reason we have developed a CLC system in dynamically operated packed bed reactors. Our novel packed bed system can be relatively easily applied at high temperatures and pressures, and is much easier to upscale for application in industry than fluidised bed systems.

What have been your most noteworthy advances in CLC technology?

This research began with the fundamental study of dynamically operated packed beds for CLC, because we believed this was a more appropriate method of achieving higher efficiency. Through experiments and modelling we have studied the behaviour of the oxygen carriers, the dynamic reactor operation, different reactor configurations and cycling strategies and the energy analysis of the integration of CLC reactors in integrated gasification combined cycle power plants in great detail. This investigation started from very small-scale testing and we are now at the hugely exciting stage where our technology is being demonstrated in a European project in which a Dutch company, Array Industries, is building a demo plant. This will provide a practical demonstration of the technology in a power plant setting.

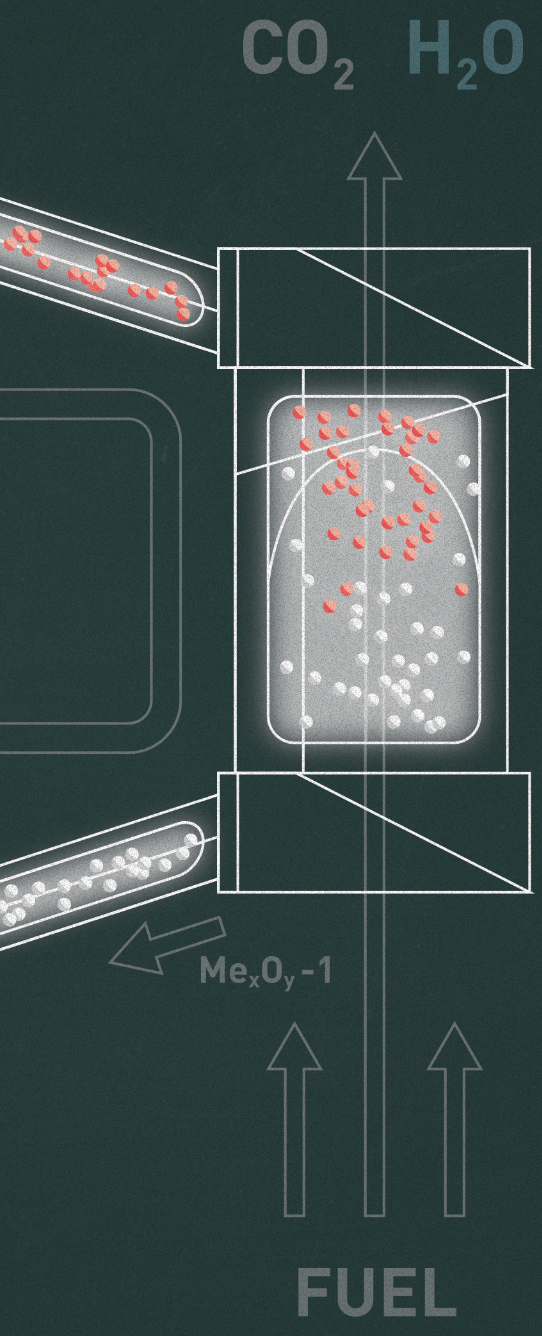


Are the recent technological advances towards the artificial production of hydrogen fuel from solar cells likely to impact on the way power plants operate?

It seems very unlikely that solar cells will replace current sources of power for the next few decades at least. Of course, hydrogen is a very good energy carrier and contributes to clean power production. However, the rising production capacity of our fossil fuel power plants will be difficult to replace with solar cells. Widespread predictions indicate that fossil fuel-based power production will continue to be predominant in the next half century. For this precise reason, carbon capture is considered enormously important in order to combat climate change.

Developing the next generation of reactors

An ongoing study at the **Eindhoven University of Technology**, Netherlands, is on the verge of creating a new form of packed bed reactor which could revolutionise fossil fuel-burning power plants



IT IS NOW widely acknowledged that anthropogenic CO_2 emissions are one of the main contributing factors to global climate change. Fossil fuel-burning power plants, the single largest source of the energy we consume, alone are responsible for an estimated 40 per cent of global CO_2 emissions. While many renewable alternatives exist, there are significant doubts over how quickly, and with what reliability, these might be upscaled to satisfy our growing demand for power. In light of this, many researchers are turning their focus towards reducing the quantities of CO_2 released by traditional power plants by developing new techniques for carbon capture and storage (CCS).

New approaches to CCS have been the subject of much research over the past decade, but many have insufficient energy efficiency. In response, a groundbreaking project based at the Eindhoven University of Technology in the Netherlands, led by Professor Martin van Sint Annaland and his colleague Dr Fausto Gallucci, is exploring the possibilities opened up by chemical looping combustion (CLC) in packed beds. Having already made great strides in their efforts to develop effective and workable solutions, there is a great deal of optimism that the research could revolutionise the capture of CO_2 from power plants, significantly reducing global carbon emissions and helping to abate the planet's changing climate.

THE BASICS OF CLC

CLC is achieved by alternately oxidising and reducing a solid metal known as an oxygen carrier (OC) via sequential contact with air and a fuel stream. The technique requires the use of a fuel reactor and an air reactor operating in a loop, with the fuel reacting with an oxidative metal oxide in the fuel reactor, transferring the oxygen from the solid to the fuel phase in order to produce CO_2 and water. In the air reactor, an air stream is then fed in so that the metal oxide is oxidised to its original form. In recent years, an increasing number of research projects have attempted to garner a clearer understanding of direct solid CLC so that existing interconnected fluidised bed reactor technology can be applied to the use of solid fuels such as coal, petroleum coke and biomass.

PACKED BED VERSUS FLUIDISED REACTORS

In terms of CLC reactor design, either a fluidised or packed bed variant can be employed. A fluidised system enables smaller solid particles to be used, circulating between the fuel and air reactors in a steady state and producing a stable stream of hot gas. However, the disadvantage of this technique is the difficulty in separating the gas and the solids, often leading to damage of the downstream gas turbines.

Van Sint Annaland advocates a more complex, but ultimately more rewarding, packed bed design. In such a reactor, larger particles are used with a gas switching system that flips between the fuel stream and the air stream for regenerating the OC. A key advantage of this approach is that although bigger particles are used, necessitating a good thermal and mechanical stability, they remain stationary in the bed, making it easier to achieve the desired pressure. Traditionally, the OC material was the subject of most research, but van Sint Annaland and his group have led the change in viewpoint. To improve CLCs, it was necessary to rethink the reactor system as well as the OC and ensure that it was capable of operating at high temperatures and pressures.

IDENTIFYING THE IDEAL OC

The most common materials used to carry oxygen in CLCs are nickel (Ni), copper, iron and manganese. As van Sint Annaland explains, a key factor in determining the suitability of an OC is how much fuel it allows to be converted into CO_2 and H_2O: "In cases of fuel slip, where not all the reactants have reacted, efficiency is decreased and some downstream cleaning processes might also be necessary". In light of this, a selectivity, ie. the amount of desired

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INTELLIGENCE

NOVEL INTEGRATED REACTORS

OBJECTIVE

To develop novel integrated, multifunctional reactor concepts based on improved fundamental understanding of multiphase reactive systems combining advanced multiscale numerical models, novel non-invasive measurement techniques and experimental demonstration.

KEY COLLABORATORS

Dr Alfredo Pacheco Tanaka, Tecnalia, Spain • **Dr Paul Cobden**, Energy Centre Netherlands (ECN), Netherlands • **Professor Giuseppe Lozza**; **Professor Paolo Chiesa**, Politecnico di Milano, Italy • **Professor Rufino Navarro**; **Professor Carlos Abanades**, CSIC, Spain • **Dr Rob Ernst**, Array Industries, Netherlands • **Dr Shahriar Amini**, SINTEF, Norway

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MARTIN VAN SINT ANNALAND obtained his PhD in 2000, and then was first Assistant Professor and later Associate Professor at Twente University in the Netherlands. He became Full Professor in 2010 and has chaired the research group Chemical Process Intensification at Eindhoven University of Technology since then. He has published more than 140 papers and been involved in three patents in the field of novel reactors and sustainable energy.

product made per reactant, as close as possible to 100 per cent is required, enabling as much CO and H_2 as possible to be converted into CO_2 and H_2O.

Ni-based materials are the most commonly studied OC and are fast-reacting and withstand very high temperatures. However, their selectivity at higher temperatures is lower than some alternatives, meaning some of the fuel remains unconverted. Unlike nickel, a copper-based OC achieves 100 per cent selectivity in all conditions but, due to its low melting point at 1,085 °C, the maximum operating temperature is relatively low, resulting in lower overall process efficiencies when using copper as the OC, at least in single-stage reactors. Iron and manganese, the two other most common OCs, can both be present in several phases, with manganese either as Mn_2O_3, Mn_3O_4, MnO and Mn. However, Mn_2O_3 breaks down at approximately 950 °C, and MnO has relatively poor mechanical stability and low oxygen transport capacity. Finally, although iron is cheap and non-toxic, and can be present as Fe_2O_3, Fe_3O_4, FeO or Fe, selectivity is a big problem, often dropping to unacceptably low values.

A COMBINATIONAL SOLUTION

As an OC that combines all the desired properties has yet to be found or designed, van Sint Annaland and his fellow researchers have developed an innovative approach to making the CLC process as efficient, cost-effective and environmentally friendly as possible. Given that the chief obstacle has been attempting to combine mechanical and chemical stability at very high temperatures with high redox kinetics at low temperatures, the team has developed a cutting-edge two-stage packed bed CLC system. By using a different OC in each of the two stages, it has become possible to combine their desired properties without designing an elusive and potentially costly new carrier. This novel two-stage variant has unlocked new doors for CLC, demonstrating that shortcomings in the properties of individual OCs need not hold back the entire development process.

THE FUTURE OF CLC

CLC technologies have yet to progress from the demonstration stage and a big step will be the integration of CLC reactors into power plants. With the building of van Sint Annaland's planned CLC reactor-containing power plant, in cooperation with, among others, Array Industries, ECN and SINTEF, a turning point will have been reached in the development of CLC technologies. The researchers at Eindhoven are convinced that, should the full potential of CLC be realised, the technology will make an enormous contribution to the implementation of CCS strategy. While conquering CCS alone does not equate to solving global climate change, it has the potential to alleviate the anthropogenic contribution to emissions by as much as 20 per cent.

Van Sint Annaland acknowledges that his research is a small, but important, piece of a much larger jigsaw: "Of course, other actions are also needed in the fight against climate change; the development of renewable energies and the improvement of energy efficiency are just two areas which will also have to progress". The research at Eindhoven is making encouraging advances towards its goal and, given the potential for CLC to expand into other fields such as chemical production, there remains a great deal of hope that anthropogenic CO_2 emissions and their resulting damage to the environment can begin to be curtailed.