Reacting to Emissions

Fluids simulation helps to speed up research into chemical-looping combustion capable of reducing fossil fuel emissions.

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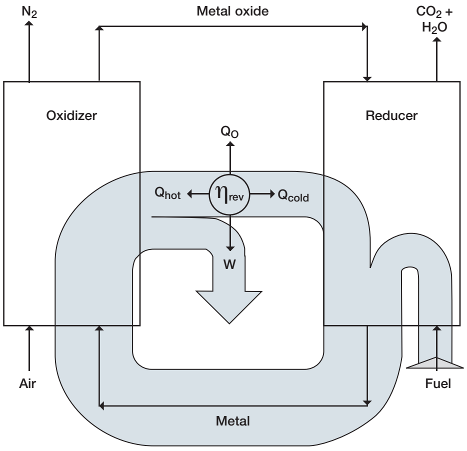
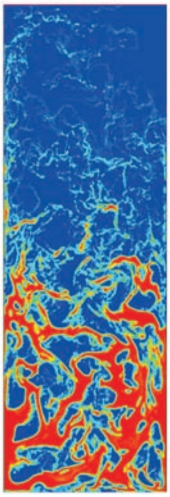


Chemical-looping combustion process

The need to reduce CO_2 emissions to restrain climate change has never been more urgent. Anthropogenic CO_2 is mainly generated in the combustion of fossil fuels, and these fuels are expected to provide a very large percentage of overall world energy consumption for the next several decades. One thing is clear: Emissions must be lowered. An accepted solution is to separate and sequester the CO_2 emitted by fuel combustion. Chemical-looping combustion (CLC) is one of the most promising technologies to carry out CO_2 capture at a low cost.

CLC is a combustion process that avoids direct mixing of fuel and combustion air. The method uses two fluidized bed reactors and circulating metal oxide that is oxidized in an air reactor and reduced in a fuel reactor to provide the oxygen required for the fuel. Pure CO_2 is obtained in the fuel reactor exhaust stream after condensation of water without the need for further gas separation. CLC will achieve significant CO_2 capture at a reduced cost when compared to other current technologies, including post-combustion amine scrubbing.

The delay in timely commercialization of CLC technology is primarily due to lack of understanding of reactive multiphase flows in the fluidized beds used in CLC systems. Design and scaleup of CLC reactors are very complex. Advanced modeling techniques are required to capture the intricate coupling between the complex reactor hydrodynamics, heterogeneous reaction kinetics and heat transfer.



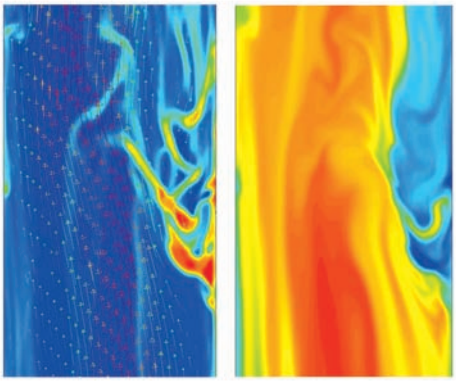
Simulation shows both the bubble (bottom half) and cluster (top half) structures occurring in fluidized beds.

With the advent of increased computational capabilities, the fundamental modeling framework of computational fluid dynamics (CFD) is emerging as a very promising additional tool for modeling reactive multiphase transport. Once the model has been validated, CFD can be used for accurate design and scaleup of the process. A team at the Flow Technology group at SINTEF, the largest independent research organization in Scandinavia, has been using ANSYS FLUENT software for this purpose in a project led by Shahriar Amini. The well-established granular flow modeling framework available within the software has been central to the progress made in this project. Fundamental model development also was made much easier by full access to all-important models by employing user-defined functions (UDF) compiled within the stable, parallelized ANSYS FLUENT solver. Heterogeneous reactions, alternate drag laws and wall functions for dilute granular flows were implemented through UDF along with several other variables, such as adjusting pressure drop over a periodic section to maintain a constant superficial gas velocity. Studies were conducted on coarse graining, implementing filtered drag, solids viscosity and solids pressure formulations by means of UDF.

One requirement for modeling the reactive gas–solids flows in CLC processes is that the software predicts formation of the particle substructures (clusters) that often occur in these systems. These structures appear as gas bubbles in dense regions and particle clusters in dilute regions of the fluidized bed reactor, and they influence everything happening in the reactor. Fine meshes and small time steps are required to resolve particle structures, but a price in computational time has to be paid to capture the physics of the system.

These particle structures increase the slip between particles and fluidizing gas. This effect is analogous to the clustering of tiny mist droplets into larger raindrops. A mist of microscopic droplets does not fall and is very easily blown away by the wind, whereas a big raindrop certainly falls with considerable force. The same happens in a fluidized bed reactor: The tiny particles conglomerate into larger clusters and, therefore, fall much more readily. If this clustering is not modeled correctly, the simulation will predict a bed behaving like a mist, but it should actually behave like a thundershower. Clearly, this will lead to substantial errors.

In addition, it is believed that particle structures influence reactions. High reaction rates are observed in regions with high volume fractions of particles and reacting gas concentrations. A dense particle cluster represents a region in which lots of surface area is available for reaction, and this significantly increases the reaction rate. However, the consequence is that the reacting gas inside the cluster is used up much faster, thereby slowing down the reaction. Clustering is actually an unwanted phenomenon in fluidized bed reactors because it slows down the reaction rate overall by concentrating all the solids in areas with a low amount of reacting gas. Incorrect cluster modeling, therefore, will predict better, but unfortunately very wrong, reactor performance.



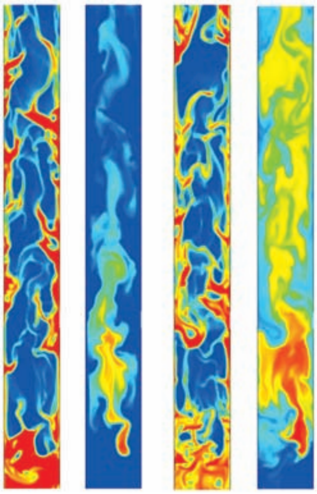
A particle cluster (left) influences the concentration of the reacting gas (right).

The team at SINTEF has accepted the challenge to contribute fundamental knowledge in the field of simulation of structure resolution on reactor performance. Results have shown that the degree to which the clusters have to be resolved (and therefore the computational cost of the simulation) depends largely on the reactivity of the particle used in the reactor. When a highly reactive particle is employed, the reaction between the fuel gas and a dense cluster is almost instantaneous and occurs almost exclusively on the surface of the cluster. In this case, the most important phenomena to be correctly modeled are the area of the cluster on which the reaction can occur and the transport of the reacting gas species to this interface. Any slight error in the position or sharpness of the cluster surface will lead to significant modeling errors.

When a less reactive particle is used in CFD simulations, however, the resolution of the cluster interface becomes less important. In this case, the reaction rate is slow enough to allow reacting gas to penetrate into the particle clusters and react throughout the reactor domain. Since the reaction occurring on the cluster surface contributes only a small amount to the global reaction rate, the correct description of this interface becomes less important.

Proper resolution of particle structures along with achieving hydrodynamic and reaction kinetic grid independence require engineers to pay particular attention to grid size compared to particle diameter. To achieve hydrodynamic grid independence for denser beds, the grid size can be 25 particle diameters or even higher, while the fine structures formed in risers need grid sizes of a maximum of 15 particle diameters, provided high-order spatial and temporal discretization is used. Reaction kinetic grid independence works the opposite way. It is very sensitive in dense beds since the volume fraction gradient on a bubble is so large. For very high reaction rates, the grid size can be 10 particle diameters or lower, whereas for 50-times lower reaction rate the grid size can be 25 particle diameters. Reaction kinetic grid independence in risers is slightly more forgiving, since the clusters can be more dilute and normally coincide with hydrodynamic grid independence.

Interestingly, simulation results show that the use of a highly reactive particle will not improve reactor performance as much as might be expected. In the highly reactive case, no reacting gas species is allowed to penetrate into the cluster. A large percentage of the particles available for reaction, therefore, is being wasted inside the dense clusters where no reacting gas species is present. When the particle with a low reactivity is used, fuel gas is available throughout the reactor, and all particles are involved in the reaction. To quantify this concept, simulations have been used to show that a 50-times decrease in particle reactivity decreases the overall reaction rate by a factor of two to three, depending on the fluidization velocity.



The interaction between particle clusters and reacting gas using a particle with high reactivity (two images on the left) and low reactivity (two images on the right). In each pair of images, the one on the left shows the particle volume fraction, and the one on the right shows the reacting gas concentration.

Information gained from this study is currently being used at SINTEF to perform fluid simulation as a design and optimization tool for CLC systems. Fundamental insights offered by these models make substantial contributions toward identifying and optimizing important design parameters to accelerate the development of this important CO_2 capture technology.