This work deals with the modeling and the direct numerical simulations of the mass transfer of a gaseous component from a gaseous phase to a liquid spherical droplet in free fall, where the absorbed component takes part to a chemical reaction. This study is realized by computing simultaneously the flow fields and the concentration fields in both gaseous and liquid phase, in order to develop a better understanding of the complex interaction between all the phenomena taking place and their effects on the transfer rate.

Keywords
Droplet, mass transfer, chemical reaction, simulation

Introduction
The absorption of gaseous materials from flue gas by reactive liquid droplets is an important mass transfer process for many engineering fields, such as air pollution control and industrial processes. One of the most studied case is the scrubbing of sulfur dioxide by a limestone slurry in a counter current spray scrubber [1,2] for reducing the emissions of SO₂.

A deep understanding of the gas-droplet mass transfer mechanisms is therefore of a fundamental interest for many applications involving sprays and a detailed modeling is essential for the good prediction of gas-liquid mass transfer coefficients. However, such a modeling is quite complex, as shown by the large number of mathematical models and experimental investigations that have been devoted to this system. Indeed, the gas-droplet mass transfer rate is influenced by several phenomena which are interacting: flows in gaseous and liquid phases, mass transport in both phases and possibly chemical reactions in liquid phase. Especially for droplet having a Reynolds number larger than 1, the flow field in the gaseous phase induces a vortex in the droplet, which can have a significant influence on the mass transfer.

This work deals with the modeling and the direct numerical simulations of the gas-droplet mass transfer of a component $A$ in gaseous phase which is absorbed by a liquid spherical droplet in free fall. A component $B$ is dissolved in the liquid droplet and this component reacts with $A$ in the liquid phase following $A + B \rightarrow C$ where $C$ is the product of the reaction.

This study is realized by computing simultaneously the flow fields and the concentration fields in both gaseous and liquid phase, which enables to describe the coupling between the phenomena. The two main objectives of this work are:
- to develop a better understanding of the phenomena governing the mass transfer rate and especially what is limiting this rate, depending on some important macroscopic parameters such as the Reynolds number and the Hatta number (which describes the chemical reaction rate);
- to compare the predictions of the model with simplified approaches in order to identify the most trustworthy and possibly to develop correlations for the use in the design of droplet absorbers.

Mathematical modeling
For water droplets smaller than 1 mm, they can be considered as rigid spheres [3]. Therefore, the proposed model involves the axisymmetric stationary motion of a non-oscillating spherical liquid droplet in free fall, at its terminal velocity, under the influence of gravity. The flow fields are computed by solving the stationary incompressible Navier-Stokes and continuity equations in both gaseous and liquid phase, imposing the continuity of the velocity and the tangential shear stress at the droplet interface.
The resistance to mass transfer is considered to occur in both liquid and gaseous phase. One convection-diffusion equation (for $A$) is considered in the gaseous phase (as there is no chemical reaction in the gaseous phase) and three convection-diffusion-reaction equations (for $A$, $B$, and $C$) are considered in the liquid phase, imposing the gas-liquid equilibrium for $A$ at the droplet interface.

The time-dependent equations of the model are solved numerically using the COMSOL Multiphysics 3.4 software, which uses the finite element method to solve the boundary-value problem. The time evolution of the mass transfer rate for various flow regimes and various chemical regimes is then studied.

**Some results and discussion**

For Reynolds number larger than unity, the presence of the vortex in the droplet and its influence on the concentration field can be observed. The case of physical absorption (without reaction) is shown in Fig. 1 for Reynolds equals 60.

It can be observed that the absorbed component $A$ is firstly convected in the external zone of the vortex, which increases the mass transfer rate. $A$ starts to fill the vortex internal zone after the saturation of the external zone. This phenomenon tends to decrease the rate of the mass transfer compared to the first step.

The transition between these two regimes can be clearly observed in Fig. 2, which presents the time evolution of the dimensionless mass transfer rate (described by the time-averaged Sherwood number) as a function of the dimensionless time (described by the Fourier number), for the case of physical absorption with various Reynolds number. A sudden change in the curve is observed for the case with a Reynolds number higher than one.

The effect on the mass transfer rate of a chemical reaction, with a moderate reaction rate (expressed by a Hatta number equals 1) and with an initial $B$ concentration approximately ten times larger than the $A$ concentration at the interface, is presented in Fig. 3.

It is observed that the chemical reaction enhances considerably the mass transfer rate. Moreover, it affects also the time of the transition between the two regimes.

This result tends to show the importance of taking into account all the phenomena simultaneously in the mass transfer modeling.

**References**

