High Temperature Process Simulation : an Example in Crystal Growth

H. Rouch , O. Geoffroy*,
INOPRO
Telespace Vercors,
118, chemin des Breux,
38250 Villard de Lans
* e-mail: olivier.geoffroy@inopro.com

Abstract: High temperature processes are used in a large variety of industrial application. Simulation help to solve technological problems and increase energy efficiency in case of industrial scale simulation. We present in this paper a research equipment simulation. The aim is to increase knowledge of temperature field in the crystal growth region in order to give researcher some important information for better interpretation of their experiments. All type of heat transfers are simulated with a great care to sensibility studies and experimental comparisons.

Keywords: high temperature thermal modeling, radiation, free convection, free surface flow, numerical design of experiments.

1. Introduction

As last year we present here an example of high temperature process simulation. Two of the main technological problems of industry using high temperature processes are the knowledge of heat gradients (for example in case of solidification, crystal growth…), or temperature prediction in all parts of an equipment (for example for design purpose). In both cases the simplest way is an analytical estimation, but usually with precision lack. In the opposite side, numerical simulations tacking in account all physical and chemical phenomenon may be more precise but is still a research topics, and still needs very large computers and long calculations. The very general procedure we follow in order to propose concrete and efficient studies is a mixed approach of a predictive use of validated models and sensitivity studies on the uncertain models and parameters. Comparison with known temperatures are performed to improve precision an decrease sensitivity studies effort.

This numerical DOE (Design Of Experiment) procedure [1] has been used in many field, for example in microwave simulation [2], and for a lot of other applications and research [3]. The example presented in this article deals with crystal growth in a reactor heated by resistors.

2. Models

2.1 Models

As mentioned in introduction the goal of this study isn't solidification modeling, but "only" a thermal characterization of the reactor during the liquid phase, with some precision on the crystal during it's melting and beginning of solidification. In this example, the heat source is not simulated, but we impose know thermal flux on the resistor side. What we need to help industrial applications is not the heating part of the process, but the temperature field near and in the crucible. This is possible because our industrial collaboration allow us to validate the complete model by comparison to experimental datas of tow types:

- Temperature profile
- Total heating power injected in the process.

Following this simplified approach the solved models are:

- Mass and momentum conservation,
- Energy conservation taking in account heat transfer by conduction, gaseous free convection, liquid free convection, and radiation,
- PDE for the P1 model

The free convection in gas is laminar, so we use the standard equations from COMSOL MULTIPHYSICS "weakly compressible Navier stockes (chns)" for this. Heat transfert including radiation is solved using "General heat transfer (htgh)". Free convection in liquid, which is also a free surface flow, is solved using the turbulence model k-epsilon. In fact, the Reynols number is between 1500 and 3000, thus the flow is not laminar, and a turbulence modeling was used, with some extra care at boundary in order to minimize errors. The liquid has also a free surface, so we refine the mesh, and use the GLS stabilization technique.

The Rayleigh number where near 1e5, which mean one recirculation may take place in the crucible, but sometimes the values grow up to 1e6, leading to a double transient recirculation with complex flow. In this case, no transient simulation where done.

The heat transfer use also participating media in the liquid phase. In this case, we use the P1 approximation, which is a compromise between accuracy and ease of implementation. This model is only valid for "optically thick" media, which is the case with the liquid.

2.2 Methodologies

The main purpose was an extensive studies of sensibility of materials properties and process conditions using numerical Design of experiments technique (DOE). The main advantage of DOE is that we can get all the interaction involved in our response, with minimum calculation, and then looking for optimization or inverse physics process. We have used a D optimal plan with quadratic model.

The main factor was some unknown or not well know emissivity, absorption coefficient in the participation media, and the repartition of the heating source. A total of 10 factor where finally considered.

We check in response different thermal gradient and temperature, in order to validate the simulation, and mainly to measure response sensitivity.

We use the parametric segregated solver for all the run involved.

2.3 Geometry, mesh, and BC

The reactor is cylindrical then we use the 2D axi-symmetrical form of the models.

The mesh is made of quadrangle for more precision and quality in all regions, which give a total of 600 000 degrees of freedom.

The boundary conditions for convective gas and liquid where wall, symmetry plane on the axis, and symmetry on the free surface. We use point setting to set up the reference pressure in the domain.

For the thermal part, surface to surface radiation where used, with some heat source where appropriate.

3. Results

3.1 Sensibility

Due to radiative transfer at high temperature, the solved system is highly non linear. The convergence strategy is the following:

We alternate flow computation and heat transfer with participative equation (P1 model) in a segregated fashion, with some relaxation for the flow equation.

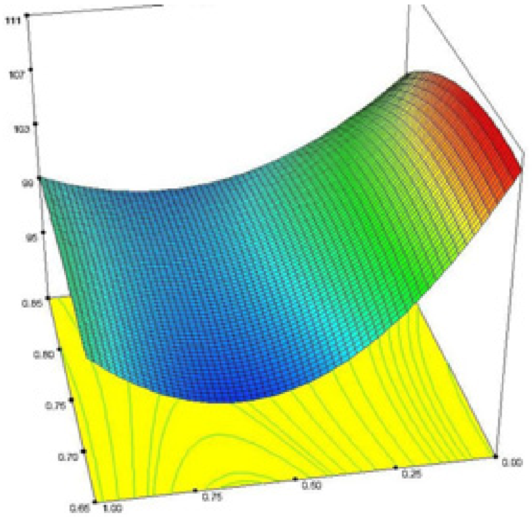

Illustration 1: Normalized thermal gradient, function of two emissivities. The objective is a gradient of 100.

A set of 75 calculations where done, and then a response surface where calculated. For all the thermal response, a very good corrected R squared close to 1 where obtained. With the quadratic equation for each response, we can look at the most influent factor for the thermal gradients near the crucible. We can see on illustration 1 a normalized gradient of temperature, and the sensitivity with two other factors : two emissivities. In this article we normalize the results to the experimental measurements: the experimental value of the same gradient is normalize to 100. In this case, we can see that for the abscissa factor, a limited set of value are valid near 0.45. For lower value, the gradient will quickly be much bigger, but for bigger value, the gradient will reach a stable place until an emissivity of 1.

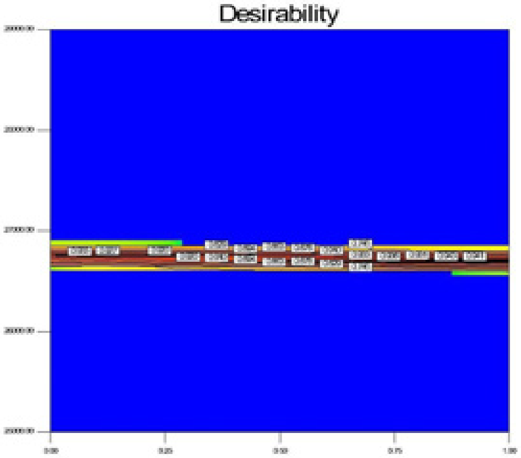

Illustration 2: Evidence that one emissivity coefficient and the process parameter are not dependent.

On the contrary, we can see on Illustration 2 an evidence of no interaction between two parameters. This illustration shows the desirability, which is a set of constraint we put on the responses, here final temperature and gradients. A desirability of 1 mean all the constraint are respected. In this case, the desirability is shown for an emissivity in abscissa and a process parameter. We can see that only a small domain for a process parameters can give satisfaction, but the solution is valid value between 0 and 1 for the emissivity, which mean no sensibility to this factor. In this particular case a simple sensibility procedure may give the same results. But this is not the case when more complex coupling between parameters exists.

3.2 Inverse simulations

For some parameters, and when we have experimental measurements, inverse physics can be done.

In this case, we can constraint the response factor (temperature and gradient), to meet the measure, even with unknown value of emissivity or absorption coefficient. In order to give good results, we need enough measurements, to be able to discriminate the sensibility to each factor. In the illustration 3, the abscissa factor (emissivity) was know to be near 0.75, but the other wasn't known. With the desirability function, we are able to find a probable value of emissivity near 0.3 +- 0.1.

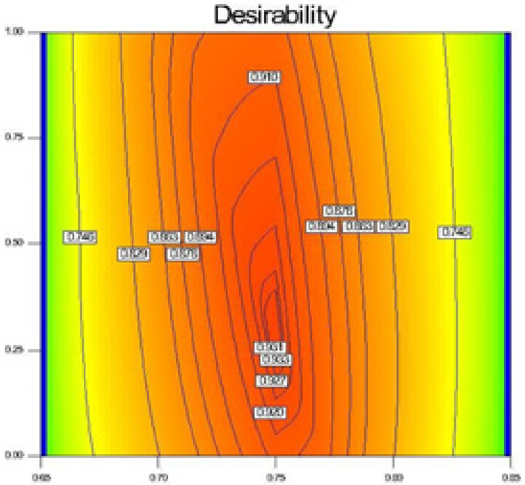

Illustration 3: Sensibility of emissivity of 2 materials, one with already know value (e.g between 0.7 and 0.8), and one unknown, which allow to use the DOE to do inverse modeling.

4. Conclusions

The crystal growth industry usually use high temperature process. To improve production efficiency, as well as research team interpretation and modeling capacities, a good knowledge of thermal effects is necessary. We show in this article a methodology to get reliable data by using numerical DOE methods for parameters adjustments and finally model validation. This allow to improve knowledge and design capacities without too complex models and computation,s and to decrease experimental plan size by predictively modeling well known phenomenon. The precision is improved by increasing comparison with experimental measurements.

5. References

1. M. Vigier, “Pratique des plans d'experiences, méthodologie Taguchi”*, Les éditions de l'organisation (1991)*

2. *H. Rouch, O. Geoffroy, “3D EM Simulation for microwave plasma coupling”, COMSOL Conference 2007.*
3. E. Gauthier, P.X. Thivel, F. Delpech, J.C. Roux, P. Ozil “An Adsorption and Photocatalysis Study of Ethyl Hexanoate”, *International Journal of Chemical Reactor Engineering,* vol 6 (2008)