A computational model for the Si/C/N nanopowder synthesis by laser pyrolysis

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Summary A numerical model was developed for the ceramic powder synthesis by the laser pyrolysis, in order to study the experiment feasibility and to optimise the synthesis process. This model takes into account heat and mass transfers during the experiment.

INTRODUCTION

For manufacturing nanocomposite ceramics, the synthesis of powder with nanometeric scale, is a promising candidate for structural applications allowing higher temperature values than those for metallic super-alloys.
This synthesis was performed by CVD, arc-plasma-heated or laser pyrolysis processes [1, 2]. The last process involves the interaction between an infrared laser beam and a gas precursor or a liquid in aerosol form. The principle is based on the resonance phenomena between the emitted laser frequency (continuous CO_2, λ=10.59 μm) with at least one of the reactant band absorption. The powder obtained by this method shows very interesting characteristics, the small reaction volume and the ability to maintain steep temperature gradients allows an accurate control of the nucleation rate, that leads to the formation of fine particles.
The control of these powders during their synthesis requires the understanding of coupled physical and chemical phenomena. Indeed, the nucleation and the growth of the particles depend directly on the resulting temperature field, dynamic field and particles concentration. These parameters are related to the operating conditions including conductive, convective and radiative heat transfers. The goal of the present work is to develop a three dimensional model of the reactor and to investigate the influence of the various thermal parameters involved during the process. The validation and the qualification at this stage are essential for any extension of the particles growth.

MODEL

Except Akmandor work at the beginning of the eighties [3], the main part of the published researches are focused on the development of the experimental part of the process. The author built a simplified model, incorporating the gas dynamics and the chemical reactions. The simulations were limited in the fact that, the thermal heat transfer did not take into account the mixture between the carrying and the reactive gas. This later becomes very important in the cooling processes of the reactive gas. The simulations were limited to a 2D-axisymmetrical geometry on a domain located around the jet, whereas the real experiment is three-dimensional and the powder synthesis may be influenced by supposed minor phenomena taking place in the full reactor in the Akmondor calculations.
In order to understand the solid particles formation mechanism and the relations which exist between experimental parameters (laser power, pressure, etc) and resulting powders characteristics, three-dimensional simulations are conducted.
Nowadays, powerful calculation means are developed in such a way that complex geometries are taken into account, incorporating the deep analysis of the physical phenomena. An industrial Computational Fluid Dynamic (CFD) package is used [4]; this software allows the modelling of various fluid flows in complex geometries area and particularly including reactive flow and permitting the incorporation of specific developed routines.

A model was developed to represent the laser pyrolysis synthesis. Steady state solution of fluid flow is commonly considered as a required condition to have a good reproducibility of the synthesis process and especially for the powder characteristics.
As a first step, the energy loss of the system by the particles thermal radiation is not taken into account. Moreover, the system incorporating the chemical species and the solid particles is under development.

The model consists of introducing the coupled conservation laws involving Navier-Stokes, energy and chemical species equations. The coupled system of Partial Differential Equations is formulated on primitive variables and solved under a finite volumes approximation on an unstructured grid.
The investigated geometry is close to the experimental configuration developed in our laboratory (figures 1, 2). It consists of the interaction between silane (SiH_4) jets surrounded by an annular argon flow (carrier gas) with an infrared laser radiation. The laser has a fixed diameter permitting the use of focused or unfocused rays.

RESULTS AND DISCUSSION

Two simulations were made to investigate the effect of the dilution of the reactant gas stream with argon that affects the heating rate. Indeed, the overall absorption coefficient is decreased, because argon does not absorb IR radiation at 10.6 µm. As seen in figure 3, the heating rate decreases and the reaction is more confined with increasing flow rate of argon.
An increase of the total pressure cell, results in an increase peak of temperature (figure 4). This indicates that, a slower velocity (velocity is inversely proportional to the density) promotes a longer residence time within the laser beam. Consequently, this leads to a higher peak the temperature.

CONCLUSIONS

A computational model for laser synthesis is developed. The temperature, dynamic, and chemical species fields are computed. These calculations are carried out in three dimensions, and take into account all thermal transfers. These results correspond to an extension of those of Akmandor's limited to a two-dimensional axisymmetric geometry and are in agreement with experimental data. Finallly, it is important to couple the local models of particles growth, with the global models of the flow to study simultaneousey the growth of the powders and their advection in the collector.

References

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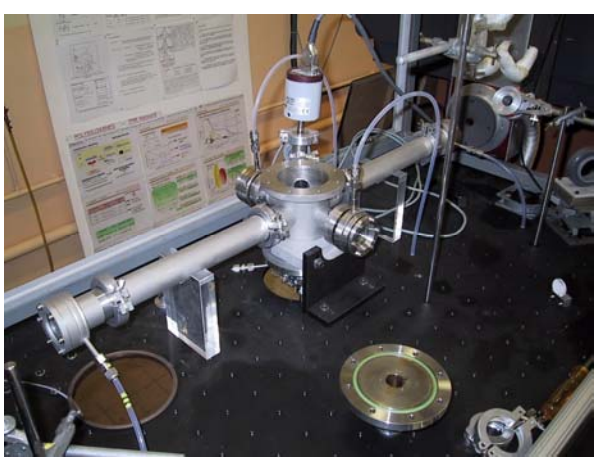

Figure 1 : Experimental setup of the nanopowder reactor

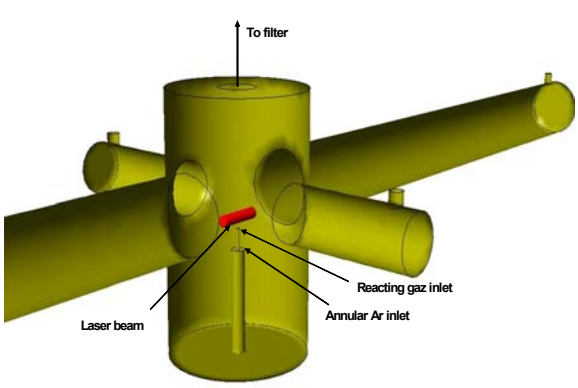

Figure 2 : Model of the reactor

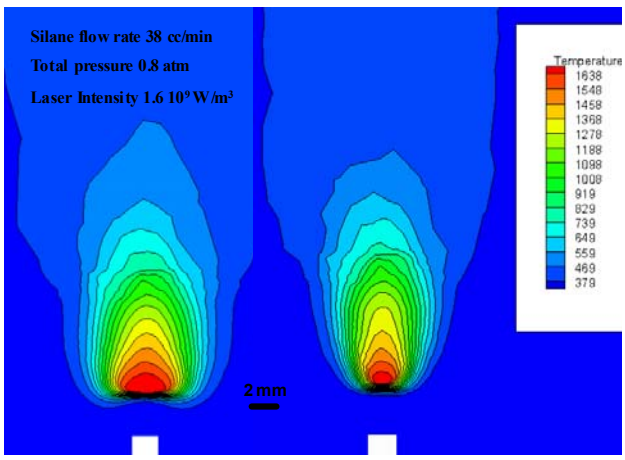

Figure 3: Temperature field for two flow rates of dilute gas STP. : 500 cm^3/min (left); 1000 cm^3/min (right).

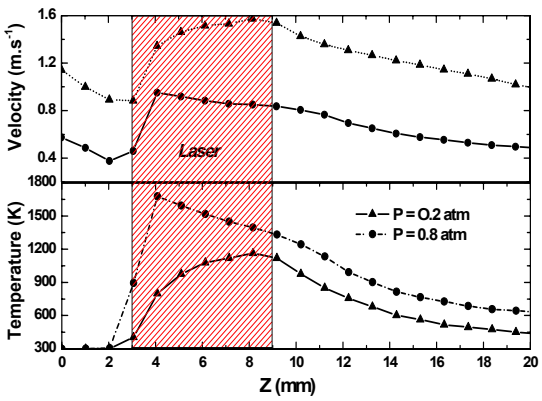

Figure 4: Maximum temperature variation and velocity, as functions of the distance above the inlet nozzle (silane flow rate 38 cm^3/min, $I_0=1.6\times10^9$ W/m^3).