

A Computational model for the Si/C/N nanopowder Synthesis by laser pyrolysis

M. Amara, D. Hourlier, M. El Ganaoui

SPCT, UMR 6638. Université de Limoges Faculté des Sciences et Techniques, 123 Albert Thomas. 87 060 Limoges

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 nanometric scale, is a promising candidate for structural applications allowing higher temperature values than those allowed by metallic super-alloys.

This synthesis was developed by CVD, arc-plasma-heated or laser pyrolysis processes [1, 2]. The last approach is a consequence of 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 CO₂, $\lambda=10.59 \mu\text{m}$) 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 allow the accurate control of the nucleation rate, permitting 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 characterize 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 majority of the published researches 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. The later is very important in the cooling processes of the reactive gas. The simulations were limited to a 2D-axisymmetrical geometry on a domain 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 Akmandor 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, and particularly including reactive flow and permits 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. The system incorporating the chemical species and the solid particles is under development.

The model consists on 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 unstructured grid.

The investigated geometry, is close to the experimental configuration developed in our laboratory (figures 1, 2). It consists of interaction between silane (SiH₄) jets surrounded by an annular argon flow (carrier gas) with 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 examine the effect of diluting the reactant gas stream with argon, this affects heating rate. Indeed, the overall absorption coefficient is decreased, because argon does not absorb IR radiation at $10.6\ \mu\text{m}$. Figure 3 shows that the heating decreased and the reaction was more confined, as the flow rate of argon increased. Therefore, we note in comparison between figure right and left (figure 3) that, increase argon flow rate, confine reaction zone.

An increase in total cell pressure, results in an increased peak temperature (figure 4). This indicates that, a slower velocity (velocity is inversely proportional to density); a longer residence time within the laser beam. Consequently, this leads to a higher peak 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. Finally, it is important to couple the local models of particles growth, with the global models of the flow to study simultaneously the growth of the powders and their advection in the collector.

References

- [1] Suhas V. Patankar, and Spalding, D. B., 1970, Heat and Mass Transfer in Boundary Layers, 2nd Edition, Intertext Books, London.
- [2] W. R. Cannon SCD, J. S. Haggerty, R. A. Marra. Sinterible Ceramic Powders from Laser-Driven Reactions: I, Powder Characteristics and Process Variables. *Journal of the American Ceramics Society*. 65 (7):324 (1982).
- [3] Akmandor I. S. Theoretical and Computational Models of Reacting Silane Gas Flows: laser Driven Pyrolysis of Subsonic and Supersonic Jets [PhD]. Cambridge, MIT (1985).
- [4] CFX5, developed by AEA technology. <http://www.software.aeat.com/>

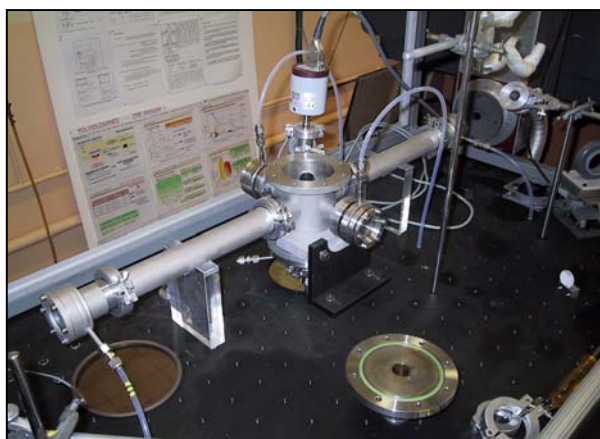


Figure 1 : Experimental setup of the nanopowder reactor

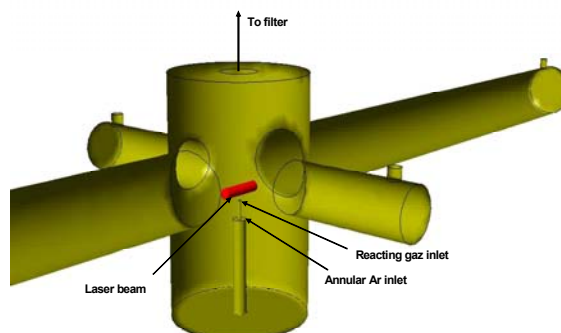


Figure 2 : Model of the reactor

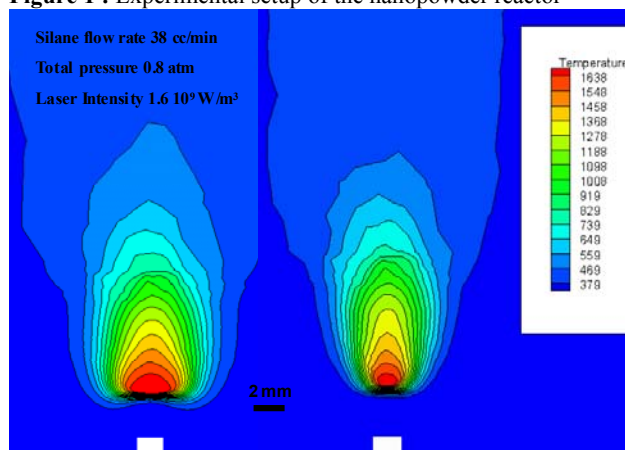


Figure 3: Temperature field for two flow rates of dilute gas STP. : $500\ \text{cm}^3/\text{min}$ (left); $1000\ \text{cm}^3/\text{min}$ (right).

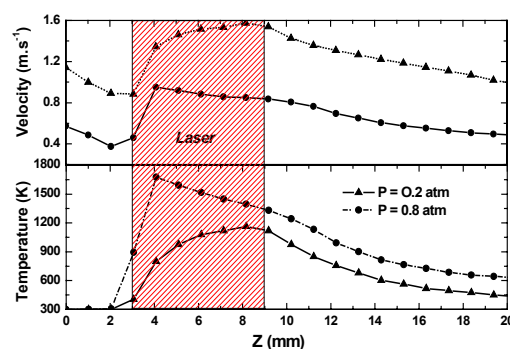


Figure 4: Maximum temperature variation and velocity, as functions of the distance above the inlet nozzle (silane flow rate $38\ \text{cm}^3/\text{min}$, $I_0=1.6 \times 10^9\ \text{W}/\text{m}^3$).