Validation of 3D modelling of an inductively coupled thermal plasma reactor through enthalpy probe measurements

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Abstract: A radio-frequency inductively coupled plasma (ICP) system for synthesis of nano-powders has been fully characterized using 2D/3D modelling. The latter has been validated through comparison with enthalpy probe measurements and the standard k-ε model with a modified source term has been found to best agree with experimental enthalpy profiles.

Keywords: thermal plasmas, ICP, nanopowders, modelling, enthalpy probe

1. Introduction

Radio frequency inductively coupled thermal plasma systems (ICP), starting with the pioneer work of Yoshida et al. [1], have been successfully used in the past 30 years to produce nanopowders of different metallic and ceramic materials. In these processes, the production of nanopowder is achieved evaporating micro-scale powders and subsequently condensing the supersaturated vapour phase. It is therefore important for tailoring the properties of the nanoparticles to control the heat transfer between the plasma and the injected precursor powders, as well as residence time and trajectories of the produced nanopowders, thus avoiding recirculating patterns in the reaction chamber [2,3].

The knowledge of the gas temperature and velocity fields is of prime importance for thermal plasma synthesis of nanoparticles because plasma properties influence the nanopowders properties, like their size distribution and composition, and the process characteristics, like deposition to the walls and yield. Therefore, the characterization of the plasma properties within the whole synthesis zone is a prerequisite for producing engineered nanoparticles with specific properties in a reproducible way. Modelling tools for thermal plasma fluid dynamics have been developed in the past twenty years and nowadays they can be used in design activities to successfully optimize industrial plasma devices [4].

Several models for the nanopowder production process in thermal plasmas have been developed and implemented in thermal plasma fluid models, allowing a deeper understanding of the process. Most advanced models take into account plasma thermo-fluid-dynamics (including electromagnetic fields and turbulence), trajectories, thermal histories and evaporation of the precursor, loading effects caused by plasma-particle interaction, vapour diffusion and, finally, nanoparticle nucleation and growth [5].

However, only few studies have been reported in literature for the validation of thermal plasma fluid-dynamic models of RF torches [6-8], and this is especially true for turbulent flows in ICP reaction chambers.

The aim of this work is to validate the developed thermal plasma model by comparison with enthalpy probe measurements. The main disadvantage of this technique is that it is an intrusive measurement. For the purpose of checking the influence of the enthalpy probe on the plasma properties 3D modelling of the ICP reactor including the enthalpy probe has been performed. The ICP setup considered in this work is the one developed by EMPA that is dedicated to the synthesis of various nanoparticles, carbides and metals among them [2,3].

This inductively coupled RF thermal plasma was characterized using a commercial enthalpy probe device (Tekna Plasma Systems, Inc.) in powder free conditions and by 2D and 3D modelling. It is shown that turbulence plays an important role in smoothing the distribution of enthalpy in the reaction chamber.

2. Enthalpy probe measurements

Enthalpy probe (EP) is an established method for assessing thermal plasma properties and is used in this work. The enthalpy probe technique has been settled to be a standard plasma diagnostic tool for the characterization of thermal plasma jets in the range from 2000 to 14000 K mainly due to its simplicity. The enthalpy measurements were performed using a commercial enthalpy probe system from Tekna Plasma System Inc. (Canada) on an inductively coupled thermal plasma reactor dedicated to nanopowder synthesis. The setup has been described already in [2] and is presented in Fig. 1. It is made of three chamber parts equipped with several viewports on which the enthalpy probe system could be adapted. The reactor inner diameter is 320 mm. The plasma torch (PL-35, Tekna Plasma Systems Inc., Canada) has been also already described in detail in [4]. A radio frequency of 13.56 MHz was used in this study. The coupling efficiency of the RF generator (Elgotec, Switzerland) was specified to be 65% of the plate power. The accuracy of the enthalpy probe measurements has been assessed (10%) and especially the deflection of the plasma plume (2 mm towards the probe tip) due to the presence of the probe has been demonstrated. The plasma enthalpy and position were measured for different process parameters like power, pressure and gas composition. Measurements have been performed along two different scan axes, using two perpendicular and two opposite viewports, as shown in figure 2.

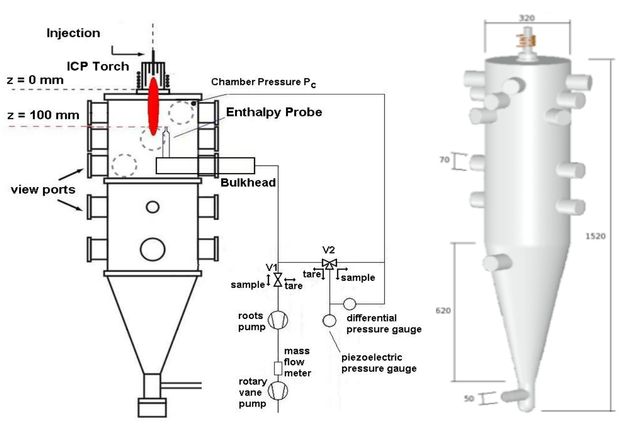

Figure 1. Schematic of the EMPA ICP system for nanopowder synthesis including the schematic of the enthalpy probe circuit (left); 3D model of the EMPA ICP system (right).

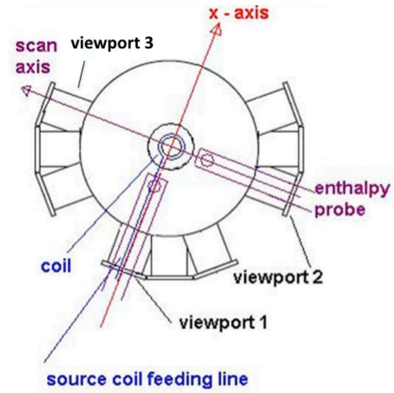

Figure 2. Schematic showing the scan axes for the enthalpy probe setup.

3. Plasma modelling

Two-dimensional simulations have been carried out in different operating conditions using a customized version of the CFD commercial code FLUENT© [4]. The governing equations can be written as

$$\nabla \cdot (\rho \vec{v}) = 0$$

$$\nabla \cdot (\rho \vec{v} \vec{v}) = -\nabla p + \nabla \cdot \tau + \rho \vec{g} + \frac{1}{2} \mathrm{Re}\left\{ \vec{J} \times \vec{B}^{*} \right\}$$

$$\nabla \cdot (\rho \vec{v} h) = -\nabla \cdot \left(\frac{k_{eff}}{c_p} \nabla h \right) + \frac{1}{2} \mathrm{Re}\left\{ \vec{J} \cdot \vec{E}^{*} \right\} - Q_r +$$

$$+\nabla \cdot \left(\sum_i h_i \left(\vec{J}_i + \frac{k}{c_p} \nabla \overline{Y}_i \right) \right)$$

where ρ is the plasma density, p is the pressure, h is the total enthalpy, v is the velocity vector, k_{eff} is the effective thermal conductivity, c_p is the specific heat at constant pressure, g is the gravitational force, E is the electric field, B is the magnetic induction, J is the current density induced in the plasma and Q_r is the volumetric radiative loss; Y_i and hi are the mass fraction and the enthalpy of the i-th gas, respectively; J_i is the mass diffusion current for the i-th gas. Diffusion of hydrogen from the sheath gas has been taken in account using the combined diffusion approach of Murphy; the electromagnetic field equations have been solved in their vector potential form using the extended field approach [4].

Since the expansion of the plasma jet from the torch to the reaction chamber induces strong flow recirculation, different turbulence models have been tested and compared with experimental enthalpy probe results. The model that best fit experimental enthalpy radial profiles at different heights turn to be the standard k-ε model, with a modified source term for the ε equation as suggested by Bolot in [9]. 3-D simulations have been performed to investigate the deflection of the plasma discharge.

Gas is supplied to the torch through three inlets with specified mass flow rates: carrier gas (4 slpm Ar) from the probe tip, primary gas (12 slpm Ar) from the inlet between the probe and the quartz tube and sheath gas (60 slpm Ar + 6 slpm H_2) from the annular gap between the quartz and ceramic tubes. The EM field has been computed using the extended field approach, thus solving the vector potential equations in a cylindrical domain around the torch (150 mm diameter, 113 mm height) on which surfaces the vector potential is assumed to vanish. In each case, coil current has been adjusted to make the total Joule power dissipated in the plasma discharge equal to the values chosen as coupled power. The operating frequency is 13,56 Mhz. The reactor is assumed to be at a pressure of 60 kPa.

3. Results and discussion

Simulations have been carried out for several operating conditions in order to fully characterize the plasma reactor for nano-powder synthesis.

In figure 2, the thermal field for two typical operating conditions (pure argon, and argon-hydrogen mixture) has been reported. The region of the plasma plume with high temperature is longer for the pure argon case, whereas in the chamber, where temperature is below the dissociation temperature of hydrogen, the two fields are very similar. Similar thermal field in the chamber arise as a consequence of very similar recirculating patterns (shown in figure 4).

Enthalpy profiles at different distances from the torch outlet have been reported in figures 5-8 for the pure argon case and argon-hydrogen case at 60 kPa, respectively. Generally, a good agreement has been found between enthalpy probe measurements and simulations. Simulations for different values of the coupled power have been carried out: for the pure argon case, the experimental values lies between the 8 kW and the 10 kW simulations; for the argon-hydrogen case, a good agreement with experimental values has been obtained with 9-10 kW of coupled power in simulations. However, independent calorimetric measurements have been done and there, a higher power has been obtained as coupled to the torch for the argon-hydrogen case (11-12 kW), leading to the conclusion that modelling slightly overestimates the enthalpy flux exiting the torch.

Turbulence plays an important role in smoothing the enthalpy profile in the plasma tail as can be seen in figures 7 and 8 where results of both the turbulent and the laminar model have been reported.

In figure 9, thermal field obtained for the argon-hydrogen case using 3D modelling has been reported. The plasma tail at 13,56 MHz is almost axi-symmetric; the thermal field in the lower part of the reaction chamber is slightly non-axisymmetric as a consequence of the recirculating patterns.

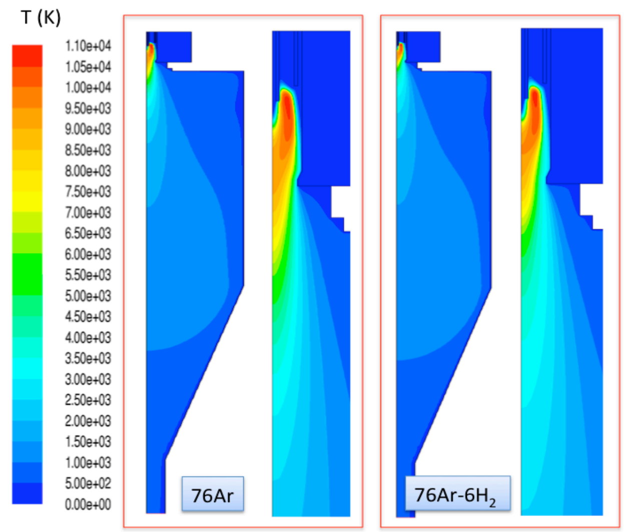

Figure 3. Temperature field in the reaction chamber (left) and detail of the torch region (right) for different plasma mixtures. Coupled power to the torch: 10 kW.

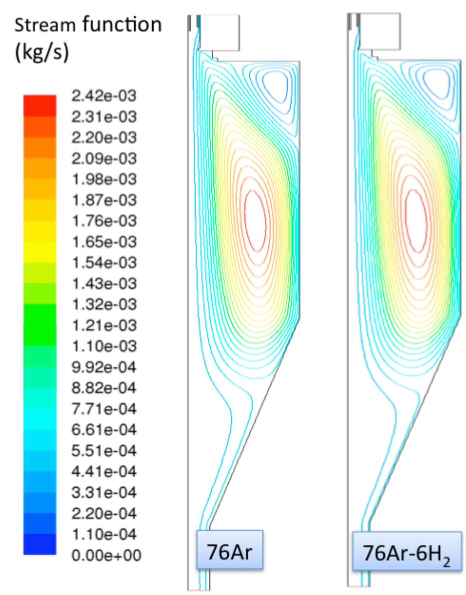

Figure 4. Stream function in the reaction chamber for different plasma mixtures. Coupled power to the torch: 10kW.

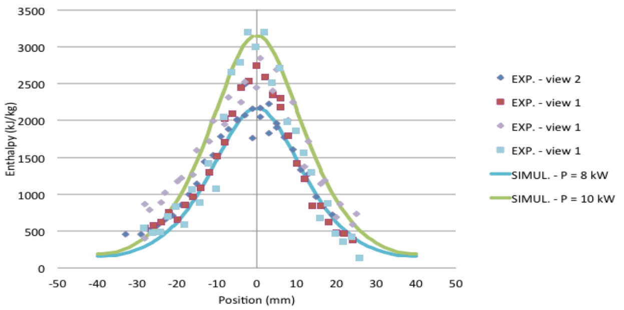

Figure 5. Comparison between enthalpy probe measurements for an argon discharge at 15 kW of plate power and modelling predictions for different values of the coupled power. Distance from the torch outlet: 60 mm.

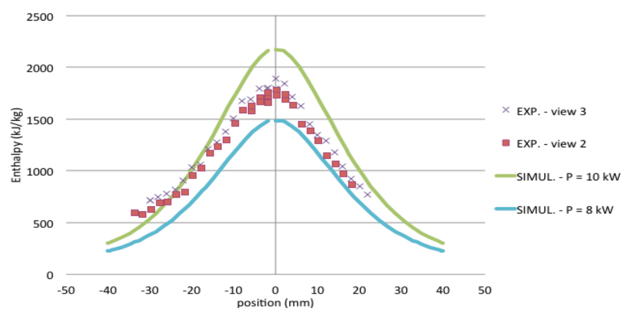

Figure 6. Comparison between enthalpy probe measurements for an argon discharge at 15 kW of plate power and modelling predictions for different values of the coupled power. Distance from the torch outlet: 100 mm.

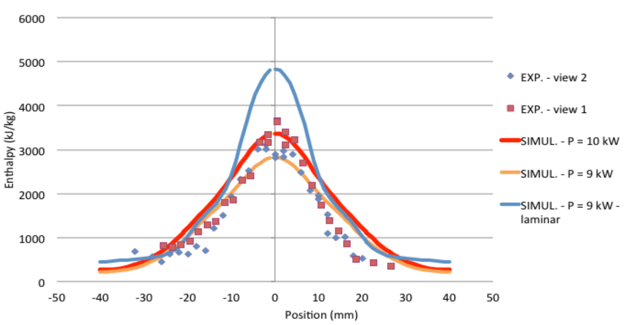

Figure 7. Comparison between enthalpy probe measurements for a 76 slpm argon – 6 slpm H_2 discharge at 18 kW of plate power and modelling predictions for different values of the coupled power. Distance from the torch outlet: 60 mm.

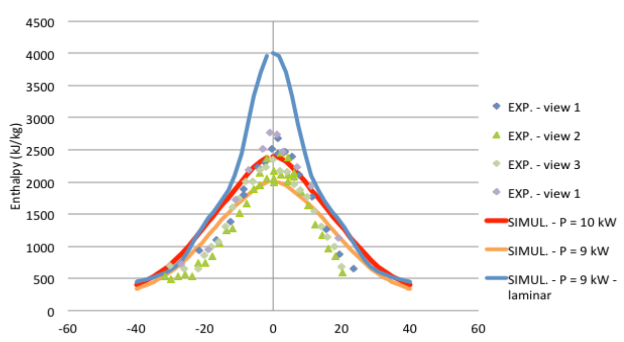

Figure 8. Comparison between enthalpy probe measurements for a 76 slpm argon – 6 slpm H_2 discharge at 18 kW of plate power and modelling predictions for different values of the coupled power. Distance from the torch outlet: 100 mm.

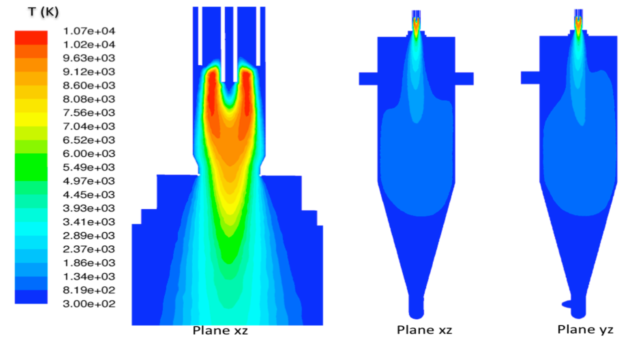

Figure 9. Temperature field in the EMPA ICP system obtained using the 3D model. Coupled power to the torch: 10 kW.

5. Conclusions

The radio-frequency inductively coupled plasma (ICP) system at EMPA for the synthesis of a wide variety of ceramic and metallic nano-powders has been fully characterized using 2D/3D modelling. The model has been validated with enthalpy probe measurements. The turbulence model that best agrees with experimental results is the standard k-ε model with the modified source term proposed by Bolot. The axi-symmetry of the discharge has been predicted by 3D modelling and verified by experimental enthalpy probe measurements.

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References

[1] Yoshida T et al. 1979 J. Mater. Sci. 14 1624-30
[2] Shin J W et al. 2006 Plasma Sources Sci. Technol. 15 441-449.
[3] Leparoux M, Schreuders C and Fauchais P 2008 Adv. Eng. Mater. 10, 1147-1150.
[4] Colombo V, Ghedini E and Sanibondi P 2010 Plasma Sources Sci. Technol. 19, 065024.
[5] Shigeta M and Watanabe T 2008 J. Appl. Phys. 103, 074903
[6] Rahmane M, Soucy G and Boulos M I 1994 Int. J. Heat Mass Transfer 37, 2035-2046
[7] Rahmane M, Soucy G and Boulos M I 1996 Plasma Chem. Plasma Process. 16, 169-189
[8] Desilets M et al. 1998 Canadian J. Chem. Eng. 76, 707-716
[9] Bolot R, Imbert M and Coddet C 2001 Int. J. Heat and Mass Trans. 44, 1095-1106