Quenching the Inductively Coupled Thermal Plasma for Nanoparticle Synthesis


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Abstract
Temperature evolution during nanoparticle synthesis by thermal plasma has an enormous influence on the final size, size distribution, and phases of the generated products. The influence of several quenching parameters, like two gas jets or eight nozzles quench ring, different gas flow rates and chemical composition, quench jet positions on the resulting temperature profiles has been investigated. The modelling has been supported and validated with experiments, in-situ diagnostics, and ex-situ measurements.

Keywords: quenching, inductively coupled plasma, nanoparticles

1. Introduction
The worldwide activities in nanoparticles research increased dramatically during the last 5 to 10 years. Among the different routes for nanoparticle production, the main directions envisaged are mechanical milling, wet chemical reactions or gas phase processes. Each of these processes has its specific advantages and limitations. Mechanical milling and wet chemical reactions are typically time intensive and batch processes, whereas gas phase processes by flames or plasma can be carried out continuously and fast. Materials of interest are mainly oxides, carbides, nitrides, and pure metals or alloys. Nano-ceramics are attractive candidates for application in coating technologies due to the expected higher coating toughness, better thermal shock and wear resistance. Inductively coupled plasma (ICP) facility provides a versatile tool for nanoparticle formation by controlling the heating, evaporation, quenching, and chemical composition of the atmosphere.

2. Experimental
The thermal plasma techniques, especially the inductively coupled plasma (ICP) process, offer many advantages compared to other gas phase techniques [1]. A high temperature and energy density enable the processing by vaporisation/condensation of high refractory materials and the use of inexpensive commercial available solid precursors [2]. Furthermore, the electrodeless operation of the ICP torch generates high purity products. The processability even under reduced pressure or reactive atmosphere offers some distinct advantages compared to other processes.

The induction plasma equipment consists of a plasma torch (PL-35, TEKNA Plasma System Inc., Canada) operated at a frequency of 13.56 MHz with an electrical input power of up to 35 kW. This torch is mounted on a modular vacuum synthesis chamber equipped with numerous view ports for in-situ monitoring. Finally the off gases are passing an on-line sampling filter or alternatively a large mass filtration unit [3] to retain the produced nanopowders. The detailed description of the experimental set-up is published elsewhere [2, 3]. A photograph of the setup is shown in Figure 1.
One important factor influencing the chemical and physical properties of the generated nanoparticles is quenching. Quenching can be carried out using different approaches e.g. using a cold surface [4], an expansion nozzle [5], or cold gas quenching [6] for nanoparticle condensation. The quench gas jets have some advantages like for instance easy positioning, variable quench gas flow rates, use of a reactive quench gas, etc. Only few references are available on this topic among them e.g. Hansen et al. [6] reporting on modelling of quench ring designs for a flame reactor or Soucy et al. [7] investigating the axial and radial mixing of two gas streams.

In this work two different quench nozzle designs are compared, i.e. a two jet nozzle and a ring type nozzle with eight holes. The influence of the different conditions on the plasma temperature and velocity field was measured by enthalpy probe technique [8]. The modelling of the temperature and velocity of the plasma flow field and quenching designs was carried out using the commercial CFD software Fluent 6.1.18. As quench gas Ar and N2 have been investigated for the simple two jet nozzles as well as for the eight nozzles quench ring. Commercially available tungsten carbide (WC) powder (d50 = 1.1µm) was used as precursor. The powder was injected into the plasma at the height of the first induction coil with a feed rate of approximately 300-350 g/min. The plasma conditions where the following (Table 1):

Table 1: Plasma parameters used for the study:

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Values</th>
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<tbody>
<tr>
<td>Central gas: Ar</td>
<td>27 slpm</td>
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<tr>
<td>Sheath gas (Ar / H2)</td>
<td>80 slpm / 6 slpm</td>
</tr>
<tr>
<td>Electrical net power</td>
<td>15 kW</td>
</tr>
<tr>
<td>Chamber pressure</td>
<td>40 kPa</td>
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<tr>
<td>Powder carrier gas: Ar</td>
<td>5 slpm</td>
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</table>
3. Results:
The symmetric two jet nozzles positioned in opposite directions have shown good agreement between experimental observations (Figure 2a) and the 3D model exhibiting the splitting of the plasma jet (Figure 2b) at an Ar quench gas flow rate of 56 slpm. The asymmetry in Figure 2a is caused by a small deviation from the symmetry axis.

Figure 2: Comparison between a) photograph of two jet quenching and b) 3D temperature modelling and c) photograph of plasma jet quenched by an eight nozzles quench ring and its modelled trend in d).

The eight nozzles quench ring performed a more homogeneous and symmetrical quenching of the hot plasma. Figure 2c) shows a photograph of the plasma at the same quench gas flow rate level (56 slpm) like the two jets in Figure 2a). The two jets nozzle intersects the plasma in two parts giving rise to uncontrolled cooling and strongly variable and broad temperature and velocity profiles. In the case of the eight nozzles quench ring, the condensed particles are forced to flow through the ring. A better control of the nanoparticle size and size distribution is expected than for the 2 jets geometry.

The influence of quenching on the resulting size of the products is shown in Figure 3. Without quenching a particle growth is observed compared to the precursor. The non-quenched product also shows a smaller size distribution due to nucleation of the vapour on the non-vaporised, i.e. cold particles. A clear reduction of the particle size is visible when quenching has been used. The size distribution depends on the temperature and velocity profiles followed by the particles.
As expected, quenching does not only influence the particle size and distribution, but also the chemistry. The XRD spectra in Figure 4 show the phase composition of plasma processed WC as a function of the quenching gas flow rate and gas chemical composition.

4. Conclusions:
The optical observations, enthalpy probe measurements [9], and CFD modelling showed all good agreements between each other. The developed CFD model allows further optimizing the quenching of the plasma by adjusting the position, the geometry and the flow rates for controlled nanoparticle synthesis.

5. Acknowledgements:
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References: