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Aspects of Heat Transfer to Particles in Thermal Plasma Processing

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ABSTRACT

Thermal plasma technology is potentially useful for a range of materials processing applications, such as the synthesis of sub-micron, ultra-pure ceramic powders. Thermal plasma reactors are characterised by short residence times (between 10 and 100 ms). Consequently, for chemical reactions to proceed to completion, reactants must be in the gas phase. Reaction rates of solids and liquids are too slow to proceed to any great degree in a thermal plasma, and unvaporised particles can contaminate product material. However, many useful reagents for plasma synthesis are available in particulate form, and thus particles must be completely vaporised if they are to be effective. In this thesis, vaporisation of particles in thermal plasmas was investigated both numerically and experimentally.

A numerical model of particle vaporisation in a thermal plasma was developed, which considers the effects of particle vapour on thermodynamic and transport properties of the plasma. This was compared with a simpler model which neglects vapour contamination effects on the plasma. Results showed that the simpler model greatly over-estimated vaporisation times of copper, aluminium, and tungsten particles in argon plasmas at temperatures less than 11000 K, but reasonable accuracy was obtained at higher temperatures. It was found that heat and mass fluxes, and vaporisation time could be expressed in a reduced form which is independent of initial particle diameter. Heat and mass fluxes during vaporisation were found to be linear functions of the inverse of particle radius. Gas-vapour property data are generally difficult to obtain, and guidelines are recommended for using pure argon properties to estimate vaporisation time.

The two major types of thermal plasma are the DC (direct current) arc, and the RF (radio-frequency), or induction, plasma. The RF plasma has several advantages over other techniques for the synthesis of powders. Reactions occur in primarily in the gas phase, resulting in good mixing between reactants. Rapid quenching of the tail flame can be used to promote homogeneous nucleation and fine particle size. There is no source of external contamination, because the RF plasma torch lacks electrodes, and a wide variety of reactants can be used, including corrosive and oxidising reagents. The plasma has a relatively low velocity and large diameter, and axial feeding of particles results in better vaporisation of particulate reagents than other thermal plasma torches.

In the experimental programme, two RF plasma torches were designed and constructed using the same 13.5 MHz, 15 kW power supply. Fluidised bed feeders and a vibratory feeder were
constructed to feed low flow rates (less than 0.2 g/min) of powders, and other apparatus were
designed for collecting product particles and quenching the plasma tail flame. The final torch
design was used to study heat transfer to particles of a range of materials and particle sizes in the
plasma. The materials studied covered a range of boiling points and heats of vaporisation, so that
the effects of these properties could be investigated.

Particles of alumina, titanium carbide and magnesium oxide smaller than 38 μm diameter were
found to vaporise completely. Condensation of vapour produced particles approximately 100 nm
diameter which were probably agglomerates of smaller particles formed by homogeneous
nucleation. Inspection of morphologies of unvaporised particles showed that the treatment of
particles in the plasma is not always uniform, as particles follow a wide range of trajectories and
experience various temperature histories. From a semi-empirical analysis of partial vaporisation
of a range of particle sizes it was estimated that the mean residence time of particles was 18 ms
and the mean plasma temperature was 9400 K. A heat transfer coefficient of 8000 W/m²K was
estimated for partially vaporising particles, which was similar to heat transfer coefficients obtained
by numerical modelling. These three parameters may be used to predict the degree of vaporisation
of particles in an RF plasma torch.

Thermodynamic analyses of plasma synthesis of titanium carbide and nitride were performed,
indicating the feasibility of the synthesis of these materials in thermal plasma reactors and possible
reactant combinations which may be used.
Acknowledgements

During the course of this project I have been fortunate enough to work with Robert Stephens, Aubrey Mathias and Anne Mette Fjellerad. Rob's contribution to the project was invaluable, and he has been a good friend and colleague. Aubrey constructed much of the apparatus and made it look like art. He also provided some handy golfing tips. Anne Mette helped with experimental runs and analysis, and her effervescence was infectious. *Jeg vil aldrig prøve at drikke som en Dansker igen!*

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List of Symbols

A  
Surface area of a particle  
Cross-sectional area of a fluidised bed  
Finite volume method coefficient  
Thomas algorithm matrix  

A_{ca}  
Cross-sectional area of plasma  

a  
Thomas algorithm coefficient  

B  
Finite volume method term  
Rotational constant  

C  
Molar concentration  

C_D  
Drag coefficient  

C_p  
Heat capacity at constant pressure  

C  
Thomas algorithm coefficient  

D  
Diffusion coefficient  
Thomas algorithm vector  

\Delta D_e  
Dissociation energy  

d  
Diameter  
Thomas algorithm term  

d_o  
Nominal particle diameter  

d_p  
Measured particle diameter  

E  
East node  

E_a  
Activation energy  

EMF  
Electromagnetic force  

E'  
Heat transfer coefficient correction factor for vaporisation  

e  
East cell interface  

F  
Force  

F_r  
Multiplicative factor for mesh generation  

f  
Frequency  

g_{eo}  
Electronic ground state degeneracy  

h  
Heat transfer coefficient  
Enthalpy  

h_{bp}  
Distance between the top of a fluidised bed and a sampling port  

\Delta h  
Enthalpy change
Ah, Heat of formation
Δh
Δh_m Heat of fusion
Δh_{ov} Overall enthalpy change
Δh_{rd} Enthalpy of decomposition
Δh_v Heat of vaporisation
h' Heat transfer coefficient corrected for vaporisation
I Heat conduction potential
Nucleation rate
Moment of inertia
Current

i' Number of molecules in a cluster of critical size
K Elutriation constant
k Thermal conductivity
Time step or time interval number
Rate constant

L Characteristic length
Plasma length

L_T Maximum allowable temperature change
M Molecular weight
m Mass
Mass of a molecule or atom

\dot{m} Mass flow rate

\dot{m}_{carrier} Mass flow rate of carrier gas
\dot{m}_{plasma} Mass flow rate of plasma gas
\dot{m}_{sheath} Mass flow rate of sheath gas in plasma-forming gases

N Molar flux
Number of turns of a solenoid
Number of nodes

P Pressure
Partial pressure
Point node
Power due to resistive (Joule) heating.

P_d Vapour pressure of a drop
P_v Vapour pressure
P_f Vapour pressure above a flat surface
P_{sat} Saturation pressure
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$P_1, P_2, P_3$</td>
<td>Finite difference coefficients</td>
</tr>
<tr>
<td>$Q$</td>
<td>Heating rate</td>
</tr>
<tr>
<td></td>
<td>Plasma discharge power</td>
</tr>
<tr>
<td>$q$</td>
<td>Heat flux</td>
</tr>
<tr>
<td>$(q)_{\text{int}}$</td>
<td>Internal partition function of an atom</td>
</tr>
<tr>
<td>$(q_\tau)_{\text{int}}$</td>
<td>Internal partition function of an ion</td>
</tr>
<tr>
<td>$R$</td>
<td>Residual resistance</td>
</tr>
<tr>
<td>$r$</td>
<td>Radius</td>
</tr>
<tr>
<td></td>
<td>Radial co-ordinate</td>
</tr>
<tr>
<td>$r_e$</td>
<td>Induction coil radius</td>
</tr>
<tr>
<td>$r_m$</td>
<td>Radius of the solid-liquid interface during melting</td>
</tr>
<tr>
<td>$r_a$</td>
<td>Plasma radius (channel model)</td>
</tr>
<tr>
<td>$r_0$</td>
<td>Initial radius</td>
</tr>
<tr>
<td>$r_9$</td>
<td>Interatomic distance</td>
</tr>
<tr>
<td>$\Delta r$</td>
<td>Cell length</td>
</tr>
<tr>
<td>$S$</td>
<td>Supersaturation ratio</td>
</tr>
<tr>
<td></td>
<td>Entropy</td>
</tr>
<tr>
<td>$T$</td>
<td>Temperature</td>
</tr>
<tr>
<td></td>
<td>Temperature vector (Thomas algorithm)</td>
</tr>
<tr>
<td>$T_b$</td>
<td>Boiling point</td>
</tr>
<tr>
<td>$T_m$</td>
<td>Melting point</td>
</tr>
<tr>
<td>$T_{\text{ref}}$</td>
<td>Reference temperature</td>
</tr>
<tr>
<td>$t$</td>
<td>Time</td>
</tr>
<tr>
<td>$t_{\text{res}}$</td>
<td>Residence time</td>
</tr>
<tr>
<td>$t_1$</td>
<td>Heating time of solid</td>
</tr>
<tr>
<td>$t_2$</td>
<td>Melting time</td>
</tr>
<tr>
<td>$t_3$</td>
<td>Heating time of liquid</td>
</tr>
<tr>
<td>$t_4$</td>
<td>Vaporisation time</td>
</tr>
<tr>
<td>$t_{\text{vap}}$</td>
<td>Total time for vaporisation, including heating and melting</td>
</tr>
<tr>
<td>$\Delta t$</td>
<td>Time step</td>
</tr>
<tr>
<td>$\Delta t_T$</td>
<td>Time interval between solutions of the energy equation</td>
</tr>
<tr>
<td>$u$</td>
<td>Velocity</td>
</tr>
<tr>
<td>$V$</td>
<td>Particle volume</td>
</tr>
<tr>
<td>$V$</td>
<td>Volumetric flow rate</td>
</tr>
<tr>
<td>$V_b$</td>
<td>Molecular volume at the boiling point</td>
</tr>
</tbody>
</table>
W  West node
    Mass of particles in a fluidised bed
w  West cell interface
    Mass of fines in a fluidised bed
X  Extent of vaporisation (%)
X_{mix}  Degree of sheath gas mixing
x  Mole fraction
y  Mass fraction
Z  Z factor
α  Under-relaxation parameter
    Condensation coefficient
Γ  Diffusivity (general)
β  Thomas algorithm term
δ  Skin depth
Thomas algorithm term
γ  Thomas algorithm term
ε  Emissivity
    Energy of molecular interaction
θ  Characteristic ionisation temperature
θ_r  Characteristic rotational temperature
θ_v  Characteristic vibrational temperature
κ  Coupling parameter
λ  Mean free path
μ  Dynamic viscosity
ν  Molar volume of liquid
ξ_o  Magnetic permeability
ρ  Density
ρ_u  Mass flux
σ  Electrical conductivity
    Lennard-Jones collision diameter
Surface tension
Symmetry number
ϕ  Magnetic flux
    Dependent variable (general)
   Thermophysical property (general)
Ω_D  Lennard-Jones collision integral for molecular diffusion
ω
Vibrational constant

Subscripts

A  Species (general)
argon  Argon
B  Species (general)
E  East node
e  East interface
Electronic
f  Mean film temperature
g  Gas (plasma)
i  Node number
l  Liquid
lim  Limit
mixture  Gas-vapour mixture
P  Point node
p  Particle
plasma  Plasma
r  Rotational
s  Particle surface
sd  Solid
t  Translational
v  Vibrational
W  West node
w  West interface
∞  Infinity (or bulk plasma)

Superscripts

-  Mean value
o  Reduced value
## Constants

<table>
<thead>
<tr>
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<tbody>
<tr>
<td>c</td>
<td>Speed of light ($= 2.99776 \times 10^8 \text{ m/s}$)</td>
</tr>
<tr>
<td>h</td>
<td>Planck constant ($= 6.6242 \times 10^{-34} \text{ Js}$)</td>
</tr>
<tr>
<td>k</td>
<td>Boltzmann constant ($= 1.38048 \times 10^{-23} \text{ J/K}$)</td>
</tr>
<tr>
<td>$m_e$</td>
<td>Mass of an electron ($= 9.1095 \times 10^{-31} \text{ kg}$)</td>
</tr>
<tr>
<td>R</td>
<td>Gas constant ($= 8.31439 \text{ J/molK}$)</td>
</tr>
<tr>
<td>$\sigma$</td>
<td>Stefan-Boltzmann constant ($= 5.67 \times 10^{-8} \text{ W/m}^2\text{K}^4$)</td>
</tr>
</tbody>
</table>

## Dimensionless groups

<table>
<thead>
<tr>
<th>Symbol</th>
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</tr>
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<tbody>
<tr>
<td>Bi</td>
<td>Biot number ($= hL/k$)</td>
</tr>
<tr>
<td>Co</td>
<td>Courant number ($= u\Delta t/\Delta x$)</td>
</tr>
<tr>
<td>Kn</td>
<td>Knudsen number ($= \lambda/L$)</td>
</tr>
<tr>
<td>Nu</td>
<td>Nusselt number ($= hd/k$)</td>
</tr>
<tr>
<td>Pr</td>
<td>Prandtl number ($= C_p\mu/k$)</td>
</tr>
<tr>
<td>Re</td>
<td>Reynolds number ($= \rho ud/\mu$)</td>
</tr>
</tbody>
</table>