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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/m^2K 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.

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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_{cs}	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
ΔD_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_{e0}	Electronic ground state degeneracy
h	Heat transfer coefficient
	Enthalpy
h_{bp}	Distance between the top of a fluidised bed and a sampling port
Δh	Enthalpy change

Δh_f	Heat of formation
Δh_m	Heat of fusion
Δh_{ov}	Overall enthalpy change
Δh_{Td}	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_o	Vapour pressure
P_s	Vapour pressure above a flat surface
P^{sat}	Saturation pressure

P_1,P_2,P_3	Finite difference coefficients
Q	Heating rate
	Plasma discharge power
q	Heat flux
$(q)_{int}$	Internal partition function of an atom
$(q_+)_{int}$	Internal partition function of an ion
R	Residual
	Resistance
r	Radius
	Radial co-ordinate
r_c	Induction coil radius
r_m	Radius of the solid-liquid interface during melting
r_n	Plasma radius (channel model)
r_o	Initial radius
r_θ	Interatomic distance
Δr	Cell length
S	Supersaturation ratio
	Entropy
T	Temperature
	Temperature vector (Thomas algorithm)
T_b	Boiling point
T_m	Melting point
T_{ref}	Reference temperature
t	Time
t_{res}	Residence time
t_1	Heating time of solid
t_2	Melting time
t_3	Heating time of liquid
t_4	Vaporisation time
t_{vap}	Total time for vaporisation, including heating and melting
Δt	Time step
Δt_T	Time interval between solutions of the energy equation
u	Velocity
V	Particle volume
$\dot{V}$	Volumetric flow rate
V_b	Molecular volume at the boiling point

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
ρu	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

c	Speed of light (= $2.99776 x 10^{8}$ m/s)
h	Planck constant (= $6.6242 x 10^{23}$ Js)
k	Boltzmann constant (= $1.38048 x 10^{-23}$ J/K)
m_e	Mass of an electron (= $9.91095 x 10^{-31}$ kg)
R	Gas constant (= 8.31439 J/molK)
σ	Stefan-Boltzmann constant (= $5.67 x 10^{-8}$ W/m^2K^4)

Dimensionless groups

Bi	Biot number (= hL/k)
Co	Courant number (= $u\Delta t/\Delta r$)
Kn	Knudsen number (= λ/L)
Nu	Nusselt number (= hd/k)
Pr	Prandtl number (= $C_p\mu/k$)
Re	Reynolds number (= $\rho ud/\mu$)