

TRANSPORT PHENOMENA DURING THE VAPORIZATION OF METALLIC  
WASTE PARTICLE IN A PLASMA JET

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ABSTRACT

Numerical simulation of the vaporization of metallic dust containing As, Pb, Zn, Sn and In is presented. The influence on the vapor composition of various phenomena such as: liquid-vapor reactive equilibrium, plasma-particle heat and mass transfer, particle temperature history is illustrated. Some spectroscopic measurements are proposed in order to validate qualitatively the model prediction tendencies.

INTRODUCTION: Generally numerical simulations concerning vaporization of particulate injected in a plasma flow deal only with some ideal cases: pure metal [1] or metalloïd [2] in a constant temperature and velocity flow. The description of more realistic situations including jet temperature and velocity profiles, complexe chemical composition of the particle [3], variation of local heat exchange due to the metallic vapors [4] is a complicated problem because of the uncertainties related, for example, to the modification of transport properties or to the high temperature chemical reactions occurring in both liquid and fluid.

This paper is an attempt for predicting the composition variation of both liquid and vapor during the vaporization of metallic dust in a plasma jet. The numerical simulation is based on a so-called global formulation of heat and mass transfer between the plasma flow and the particle.

MODEL FORMULATION : The main assumptions are the following :

- Chemical reactions are fast enough to be governed by thermodynamic equilibrium.
- The liquid is ideal.
- Plasma-particle heat and mass transfer may be described by overall coefficients.
- The flow is not modified by the particle injection.
- The vapors don't change the plasma composition far from the particle.

The structure of the model is presented by the following diagram :

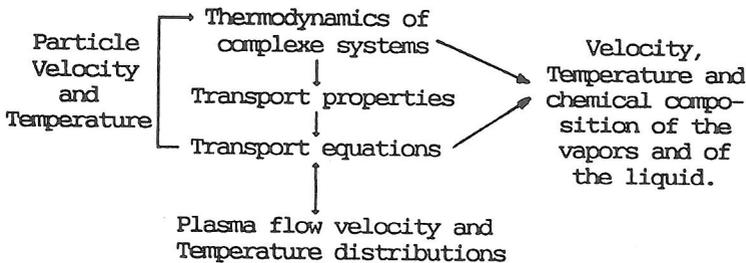
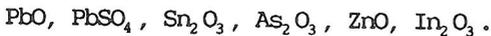


Table 1 gives the composition of the dust :

Element	Pb	Sn	As	Zn	In	S
Weight	48.5	16.5	5.8	4.2	2.2	2.5

The dust is composed of oxides and sulfate mainly :



NUMERICAL SIMULATION :

Thermodynamics : In order to calculate the equilibrium composition of the system as a function of temperature, oxygen pressure and condensed phase composition, we have taken into account the following chemical elements : Pb, Sn, As, Zn, In, S, O and Ar or Ar/H<sub>2</sub>. The effect of temperature on the equivalent vapor pressure of the chemical species is illustrated in figure 1. The equivalent vapor pressure of the element J is defined by :

$$P_j = \sum_{i=1}^N A(i,j) P_i \quad (1)$$

Figure 1 shows that the most volatile elements in argon are As, S and Pb then Zn, Sn and In. Arsenic, lead and tin are oxidized (mainly As<sub>2</sub>O<sub>3</sub>, AsO, PbO and SnO) while zinc and indium are mainly in the metallic form.

Assuming a vaporization at constant temperature T=1900K, there is a variation of the condensed phase (liquid + solid) composition during the process. This modification is illustrated in Figure 2. Due to the large vapor pressure of "Pb" the relative concentration of this element decreases rapidly as a function of mass loss and the concentration of "Sn" increases because it is more stable.

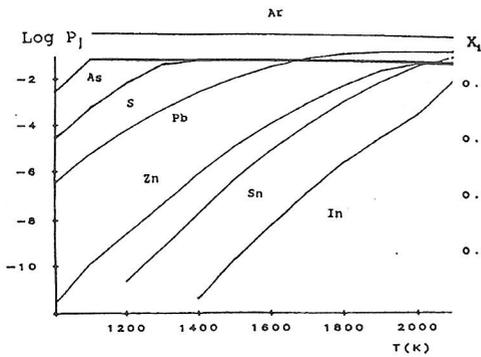


Fig.1: Equivalent vapor pressure of the elements as a function of temperature  $P_r=1$  atm,  $n_{h,r}=1$  mol.

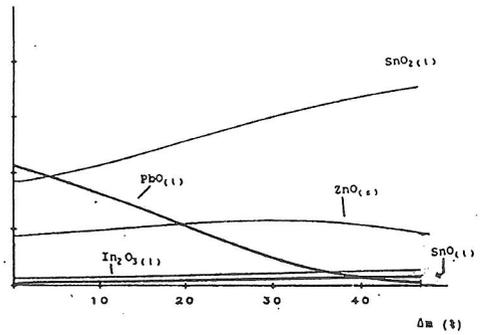


Fig.2: Variation of the condensed phase composition as a function of the mass loss at  $T=1900$  K.

Heat and Mass Transfer : Momentum and heat transfer equations are detailed in [5] as well as the mass transfer phenomena. The mass flow rate of the species  $i$  at the radius  $r$ ,  $\dot{m}_i(r)$  is given by the following equations :

$$\dot{m}_i(r) = -4\pi r^2 p \varrho_m D_{im} \frac{\partial Y_{ig}(r)}{\partial r} + Y_{ig}(r) \dot{m}_T \quad (2)$$

$$\text{with } Y_{ig}(r) = \frac{P_i(r)M_i}{N \sum_{j=1}^N P_j(r)M_j} \quad (3)$$

$$\dot{m}_T = \sum_{i=1}^{N-1} \dot{m}_i(r_p) \quad (4)$$

accounting for the boundary conditions :

$$r = r_p \quad Y_{ig}(r_p) = \frac{P_i(r_p) M_i}{N \sum_{i=1}^N P_i(r_p) M_i} \quad (5)$$

$$r = r_f \quad Y_{ig}(r_f) = Y_{ig} = 0 \quad i = 1, 2, \dots, N-1$$

$$Y_{ng}(r_f) = 1 \quad (N \text{ corresponds to Argon})$$

and with :

$$\varrho_m = \sum_{i=1}^N \frac{P_i M_i}{RT_F}, \quad T_F = [T(r_p) + T(r_f)]/2$$

$$D_{i,m} = \frac{1 - X_i}{\sum_{i=1, i \neq j}^N X_i / D_{i,j} (T_F)}, \quad X_i = [(X_i(r_p) + X_i(r_f))] / 2$$

The first term of equation (2) expresses the diffusion mass flow rate of  $i$ , it may be written as :

$$J_i(r) = Sh_i S_p \rho_m D_{i,m} [Y_{i,g}(r) - Y_{i,g}(r_f)] / d_p \quad (6)$$

Then it comes :

$$\dot{m}_i = \sum_{i=1}^{N-1} Sh_i S_p \rho_m D_{i,m} [Y_{i,g}(r) - Y_{i,g}(r_f)] / 2r_p [1 - \sum_{i=1}^{N-1} Y_{i,g}(r)] \quad (7)$$

a comparison of various formulation of the Sherwood number,  $Sh_i$ , is proposed in [5]. The correlation from [6] was chosen :

$$Sh_i = (2 + 0.87 Re_m^{1/2} Sc_F^{1/3}) (1 + EM_i)^{-0.7} \quad (8)$$

$$10 < Re_m < 2000$$

$$Re_m = \rho_f d_p |U_f - U_p| / \mu_f$$

$$EM_i = ([Y_{i,g}(r_p) - Y_{i,g}(r_f)] / [1 - Y_{i,g}(r_p)])$$

If a 100 $\mu$ m in diameter dust particle is injected in an argon flow at 10 000 K and 300 m/s, the surface temperature reaches a plateau at 2040 K and  $t \sim 2,5$  ms. The relative composition of the vapor is illustrated in Fig. 3 (As and S are supposed to be totality vaporized during the first moment). Comparing figure 3 and figure 1 it appears that mass transfer modifies strongly the vaporization tendency predicted by thermodynamics. Under this condition the selectivity of the process is very poor and Zn is more volatile than Pb. This is due to the relative value of the diffusion coefficients of the various chemical species. Typical variation of the relative mass loss of the particle with respect to plasma temperature and particle diameter was calculated, for example, a 25  $\mu$ m in diameter dust particle disappears completely after 3 ms in a 10 000 K argon plasma, but for  $d_p=100 \mu$ m this characteristic time is about 60 ms.

The particle in a plasma jet : In order to account for a realistic description of the flow are have used the analytical model of plasma jet proposed by [7]. For example the trajectory of a particle injected at 1 m/s in the axis of the jet is shown in figure 4. the variation of the relative concentration of the metallic vapors along the axis in an Ar-H<sub>2</sub> (10%) plasma jet is presented en figure 5.

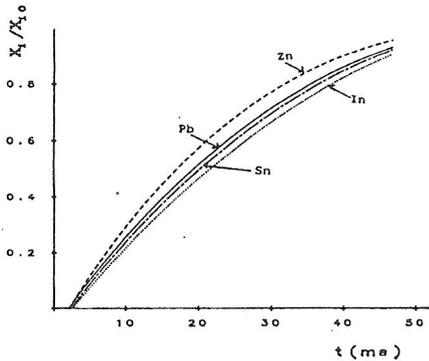


Fig. 3: Variation of the relative molar fraction of the metals in the vapor as a function of time. Argon,  $T=10\ 000\text{K}$ ,  $U_f=300\text{ m/s}$

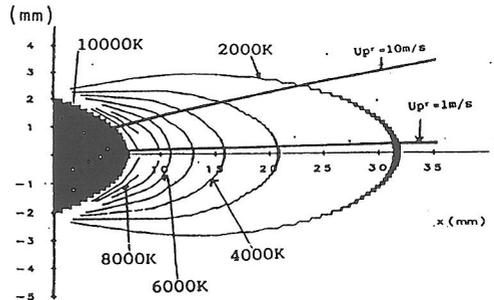


Fig. 4: Temperature profile in the argon plasma jet and particle trajectories.

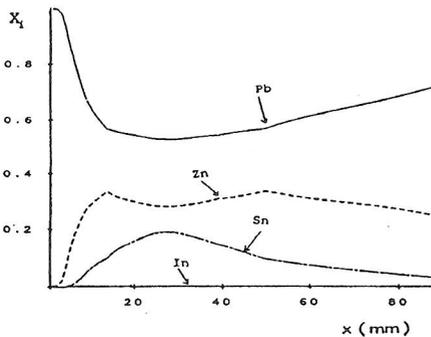


Fig. 5: Variation of the molar fraction of the metallic vapors in an  $\text{Ar-H}_2$  plasma jet.

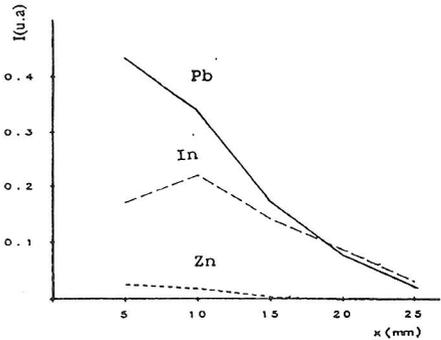


Fig. 6: Experimental results: relative intensities of the rays related to the metallic vapors along the axis of an  $\text{Ar-H}_2$  plasma.

Due to the variation of temperature along the trajectory lead is the most volatile whereas indium concentration in the gas phase is very low. This data indicates that the separation of the metallic compounds resulting from combined chemical reaction, heat and mass transfer is possible in an  $\text{Ar-H}_2$  plasma jet.

**EXPERIMENTAL STUDY** : In order to validate the prediction tendencies we have measured the atomic emission of the vapors in the plasma flow by spectroscopic analysis. The data permit two comparisons between predictions and actual phenomena: the nature of metallic elements in the vapor and the relative intensities of each. The injected particles are 80 mm agglomerates resulting from the processing of fines dust ( $dp < 5\ \mu\text{m}$ ). A typical result is shown in figure 6. First

of all, Sn was not detected in the visible range of our analysis. Second we have detected Pb, In and Zn but In vapor seems to be more abundant than Zn. This difference may be due to : (i) an overheating of the particle surface due to the uncertainty about the agglomerate particle effective thermal conductivity. (ii) a bad prediction of the influence of water vapor on thermodynamic equilibrium.

CONCLUSION : We have proposed a general model for prediction of the behavior of waste dust in a plasma flow. The numerical simulation takes into account combined momentum, heat, mass transfers with chemical reaction. A validation is now in progress including spectroscopic measurements. This approach will be a useful tool for the design of plasma processes for the treatment of metallic wastes.

Acknowledgment :

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Notation :

$A(i,j)$  : atom number of  $j$  in the gas species  $i$

$D_{i,m}$  : diffusion coefficient of  $i$  in the mixture

$d_p$  : particle diameter.

$M$  : molar mass.

$m$  : mass flowrate.

$P$  : pressure

$Re$  : Reynolds number

$r$  : radius

$S$  : surface

$Sc$  : Schmidt number,  $Sc = \frac{\mu}{\rho D}$

$Sh$  : Sherwood number,  $Sh = \frac{hd}{D}$

$T$  : temperature.

$U$  : velocity.

$X_i$  : molar fraction of  $i$ .

$Y_i$  : mass fraction of  $i$ .

$\rho$  : density.

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