

PRODUCING MONODISPERSE POWDERS FROM REFRACTORY METALS
BY GAS-PLASMA DISPERSAL

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Metal particle formation in a plasma jet is described. The conditions necessary for monodisperse production are examined.

Various liquids can be produced as monodisperse droplets [1, 2] from Rayleigh instability in a jet under mechanical perturbations, but this is not applicable to dispersing metals with melting points above 1000°C and particularly above 2000°C because of technical difficulties, namely melting of much of the metal, producing a jet with set parameters, and so on.

We consider the necessary conditions for monodisperse particle production for these two groups of high-melting metals. Those conditions for substances with melting points over 1000-2000°C include [1, 2]: high-temperature heating to liquefy the material, jet production, and the application of a constant or periodic force to produce droplets. We thus consider the scope for realizing such dispersal as a function of melting point. We consider a solid jet (wire), which is locally heated at the end by a plasma, and where the resulting liquid is subject to a constant or periodic force.

Gas-plasma liquid dispersal involves the feature that the local hot zone is produced by a plasma from the combustion of hexane, propane, acetylene, and so on in an oxidant. The forces acting on the droplets arise from the aerodynamic force from the plasma jet.

The dispersal occurs qualitatively as follows. The rod is supplied continuously at rate v_1 to the plasma, where it receives the heat required for heating and melting. After the metal melts, capillary forces result in a drop at the end of the rod. As the drop grows, it is subject to increasing gravitational and aerodynamic forces, and when the sum of them exceeds the capillary forces, the droplet detaches.

To check that monodisperse droplets can be produced in this way, one needs a strict control on parameters such as the rod speed in relation to diameter, the thermal conductivity, the temperature, the plasma speed, and so on.

We approximate the heat transfer from the jet in terms of a plane-parallel flow of hot air having Π -shaped temperature and velocity profiles. The other end of the rod is in contact with a remote body having a high thermal capacity and environmental temperature. There are two types of heat transfer: convective and radiative.

The convective transfer is evaluated from Nu for a cylinder in a transverse flow of hot air [3]:

$$Nu = c \cdot Re^n. \quad (1)$$

In our case, $c = 0.81$ and $n = 0.4$, and $Re = (v \cdot d) / \nu$ is calculated on the basis that the kinematic viscosity is $\nu = f(T)$. From Nu we determine the heat-transfer coefficient

$$\alpha_1 = Nu \lambda / d. \quad (2)$$

We determine α_1 on the basis that $\lambda = f(T)$ for air to get the heat flux to the cylinder on the assumption (as the area is small) that the temperature is everywhere the same and equal to the melting point:

TABLE 1. Total Heat Flux to Heated Part of Cylinder

$d, 10^{-4} \text{ m}$	$Q, \text{ W}$
1,4	4,4
2,4	6,4
2,8	7,1

$$Q_1 = \alpha_1 S (T - T_{\text{mp}}). \quad (3)$$

The radiation flux from the jet to the cylinder is given by Stefan's law [3]:

$$Q_2 = \varepsilon_1 \varepsilon_2 S \sigma (T^4 - T_{\text{mp}}^4). \quad (4)$$

The integral flux to the heated part of the cylinder is found by summing (3) and (4):

$$Q = \alpha_1 S (T - T_{\text{mp}}) + \sigma \varepsilon_1 \varepsilon_2 S (T^4 - T_{\text{mp}}^4). \quad (5)$$

Table 1 gives the total flux for copper rods with various diameters when the length of the part is $\ell = 6 \times 10^{-3} \text{ m}$ and the temperature is 2273 K, while the flow has $v = 10 \text{ m/sec}$, $\varepsilon_1 \approx 0.6$, $\varepsilon_2 \approx 0.85$, as determined from (3).

When one estimates the melting rate, one needs to consider not only the heat transfer from the jet but also that from the free part of the rod to the surrounding medium. We consider free convection and radiation.

There is a relation between the numbers defining the free convection:

$$\text{Nu} = C (\text{Gr} \cdot \text{Pr})^n, \quad (6)$$

in which $\text{Gr} = \frac{d^3 g \Delta T}{T_m \nu^2}$; $\text{Pr} = \nu / \chi$; $\chi = \lambda / \rho c_p$, determined for T_m . The surface temperature of the rod varies from the melting point T_{mp} at the point of contact with the heater to the environmental temperature T_{en} at infinity. The characteristic surface temperature T_{su} is the arithmetic mean of these two, $T_{\text{su}} = (T_{\text{mp}} + T_{\text{en}})/2$. The temperature difference is $T = T_{\text{su}} - T_{\text{en}} = (T_{\text{mp}} - T_{\text{en}})/2$, and T_m is the mean temperature of the boundary layer, which is

$$T_m = \frac{1}{2} (T_{\text{su}} + T_{\text{en}}) = \frac{1}{4} (T_{\text{mp}} + 3T_{\text{en}}).$$

The c and n in (6) are constants, which for Gr and Pr characteristic of the process are $c = 1.18$ and $n = 1/8$. One determines Gr and Pr to derive the α_3 related to free convection:

$$\alpha_3 = \lambda c (\text{Gr} \cdot \text{Pr})^n / d. \quad (7)$$

For q_1 we have

$$q_1 = \sigma \varepsilon_1 (T^4 - T_{\text{en}}^4), \quad (8)$$

in which T is the surface temperature at the given point. We rewrite (8) as

$$q_1 = \sigma \varepsilon_1 [(T^2 + T_{\text{en}}^2)(T + T_{\text{en}})](T - T_{\text{en}}). \quad (9)$$

The expression in square brackets on the right in (9) is defined for $T = T_{\text{su}}$ and is taken as the effective radiative transfer coefficient:

$$\alpha_4 = \sigma \varepsilon_1 (T_{\text{su}}^2 + T_{\text{en}}^2)(T_{\text{su}} + T_{\text{en}}). \quad (10)$$

TABLE 2. Relation between Wire Feed Rate and Radius

$r, 10^{-4} \text{ m}$	$Q, \text{ W}$	$v_1, 10^{-2} \text{ m/sec}$
1,4	1,9	2,02
1,2	1,57	2,57
0,7	0,8	5,28

We determine the radiative heat-transfer coefficient α_4 and the convective coefficient α_3 to get the total transfer coefficient as additive:

$$\alpha = \alpha_3 + \alpha_4. \quad (11)$$

We thus have all the necessary parameters for deriving the temperature in the free part of the rod.

We assume that the specific heat of the rod is fairly large and that the temperature does not vary over the cross section, i.e., the treatment is one-dimensional and the temperature varies only along the x coordinate.

The equation for heat transfer in the wire is [3]

$$\lambda_1 S \frac{\partial^2 T}{\partial x^2} = \alpha P (T - T_{\text{en}}) - \rho c_p S v_1 \frac{dT}{dx}. \quad (12)$$

The left-hand side is the change in the amount of heat flowing through S. The first term on the right describes the radiative-conductive heat transfer from the side surface. The second term describes the heat transfer through the cross section due to the motion at v_1 . The minus sign corresponds to the temperature decreasing along the rod, i.e., $dT/dx < 0$. As $S = \pi r^2$ and $P = 2\pi r$, we rewrite (12):

$$\partial^2 T / \partial x^2 + (\rho c_p v_1 / \lambda_1) (\partial T / \partial x) - 2\alpha T / \lambda_1 r + 2\alpha T_{\text{en}} / \lambda_1 r = 0. \quad (13)$$

We solve (13) with the boundary conditions $T = T_{\text{mp}}$ at $x = 0$ and $T \rightarrow T_{\text{en}}$ for $x \rightarrow \infty$ to get the temperature distribution:

$$T = (T - T_{\text{en}}) \exp\left(-\sqrt{\rho^2 c_p^2 v_1^2 / 4\lambda_1^2 + 2\alpha / \lambda_1 r} x + T_{\text{en}}\right). \quad (14)$$

We determine the total heat loss through the side surface, which is governed by the heat flux through the cross section at $x = 0$:

$$Q_3 = -\pi r^2 \lambda (\partial T / \partial x)_{x=0}. \quad (15)$$

We differentiate (14) with respect to x and substitute for the derivative at $x = 0$ into (15) to get

$$Q_3 = \pi r^2 \lambda_1 (T_{\text{mp}} - T_{\text{en}}) \left(\sqrt{-\rho^2 \frac{c_p^2 v_1^2}{4\lambda_1^2} + \frac{2\alpha}{\lambda_1 r}} + \rho \frac{c_p v_1}{2\lambda_1} \right). \quad (16)$$

We consider the heat balance equation for the melting of wire advancing through an immobile transition front at a constant rate:

$$Q = \rho L \pi r^2 v_1 + Q_3, \quad (17)$$

in which Q is defined by (5). The second term on the right is the power needed to melt the wire moving into the melting zone at speed v_1 . We substitute for it into (17) and solve for v_1 to get the speed as a function of the physicochemical parameters and thermal power Q from the external source:

$$v_1 = \frac{[2L(T_{mp} - T_{en})c_p]Q}{2\pi Lr^2\rho[L + (T_{mp} - T_{en})c_p]} - \frac{T_{mp} - T_{en}}{2\rho\pi r^2 L} K, \quad (18)$$

in which

$$K = \left(\frac{c_p^2 Q^2 + 8\pi^2 r^3 \alpha L \lambda [L + (T_{mp} - T_{en})c_p]}{[L + (T_{mp} - T_{en})c_p]^2} \right)^{1/2}.$$

We substitute $v_1 = 0$ into (18) to get the minimal thermal power producing melting:

$$Q = \sqrt{2\alpha\lambda_1}\pi r^{3/2} (T_{mp} - T_{en}). \quad (19)$$

We use (19) and (18) to estimate the amount of heat needed to melt rods with various radii and made of various materials, and also the feed rate as a function of cross section.

Table 2 gives Q and v_1 for copper wires and jets having $v = 10$ m/sec and $T = 2273$ K.

We thus have expressions for the amount of heat needed to melt the end of a rod for various materials with various dimensions, together with the corresponding feed rates.

We now estimate the size of the microgranules that can be made in this way. The following forces are involved: surface tension, gravitation, and aerodynamic resistance.

The radius is derived on the basis that the resistance and the gravitational force are at 90° , while their resultant and the axial component of the surface tension act along the same straight line.

The equilibrium condition defines the radius:

$$F_{st} = (m^2 g^2 + F_s^2)^{1/2}, \quad (20)$$

in which F_{st} is the axial component of the surface tension force:

$$F_{st} = 2\pi r \beta \cos \varphi, \quad (21)$$

where β is taken at T_{mp} and

$$\cos \varphi = r/r_d \quad (22)$$

(r_d is droplet radius), and

$$F_s = 6\pi\eta r_d v [1 + 0,265(vr_d/v)^{2/3}]. \quad (23)$$

We substitute (21)-(23) into (20) to determine r_d . The numerical solution to (20) with characteristic jet parameters for granules made from a copper rod having $r = 7 \cdot 10^{-5}$ m gives $r_d \approx 160$ μ m.

The parameters deived from these estimates show that the scheme can give monodisperse dropets for metals with moderate and high melting points.

NOTATION

Nu, Nusselt number; Re, Reynolds number; c, coefficient of proportionality; n, exponent; ν , kinematic viscosity; d, wire diameter; T, temperature; $\alpha_1, \alpha_2, \alpha_3, \alpha_4, \alpha$, heat-transfer coefficients; λ , thermal conductivity; T_{mp} , melting point; l , heating length; s, heating surface area; Q_1 , heat flux from hot jet to heated part; Q_2 , radiative heat flux; σ , Stefan's constant; ε_1 and ε_2 , blacknesses of cylinder and hot gas; Q, total heat flux; Gr, Grashof number; g, acceleration due to gravity; Pr, Prandtl number; ΔT , temperature difference between rod surface and environment; T_m , characteristic temperature; T_{su} , surface temperature, T_{en} , gas temperature; χ , thermal diffusivity; ρ , density; c_p , specific heat; q_1 , radiative energy flux from unit surface; x, longitudinal coordinate; S and P, area and perimeter for cross section of rod correspondingly; K_1 and K_2 , roots of characteristic equation; T_1 and T_2 , particular solutions to (13); r, rod radius; c_1 and c_2 , coefficients; Q_3 heat flux from rod; L, latent heat of fusion; v_1 , rod supply rate; K, coefficient in solution; F_{st} , axial component of surface tension force; β , surface tension coefficient; F_s , Stokes force; η , dynamic viscosity of gas; v, plasma jet speed.

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