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Inertial impaction of heavy molecules

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The transition from diffusion-dominated to inertia-dominated behaviour in the transport of condensable heavy molecules carried in a continuum subsonic He jet that impinges on a solid surface is studied experimentally. The Stokes number S, or ratio between the heavy-molecule relaxation time and the fluid-dynamic time, is varied in the interval $0 \lesssim S \lesssim 1$ by changing the jet Mach number at a constant value of the Reynolds number. Although the heavy species departs considerably from equilibrium at all but the smallest values of S, the helium jet is always near equilibrium conditions. At values of S of order unity the observed rate of deposition at the stagnation point asymptotes to a value some six times greater than in the diffusion region (where $S \rightarrow 0$), implying that the process is governed by the large inertia of the heavy species, very much like in aerosol impactors. As a result, it is argued that the concept of pressure diffusion is unsuitable to explain the observed behaviour. An approximate theoretical description of the transport process is given for the region $S \ll 1$ where the kinetic problem is amenable to a hydrodynamic treatment. Finally, the analogy with the inertia-dominated behaviour of aerosols is used to assess the relative merits of various aerodynamics schemes aiming at separating isotopes.

1. Introduction

More than 20 years ago, Reis & Fenn (1963) suggested that some of the highly unusual phenomena that had been observed in the evolution of gas mixtures with disparate molecular weights could be explained by analogy with the behaviour of aerosol particles. Such a proposition should have naturally gained wide acceptance in the field, since there is no qualitative difference between a dilute suspension of heavy molecules and one of microscopic particles.† Surprisingly, however, many of its important implications on the non-equilibrium behaviour of heavy molecules still remain insufficiently explored, and are reexamined here.

As is well known, microscopic particles are endowed with a non-negligible inertia, and are coupled to the suspending gas through a finite dragging force. They thus need a finite time τ (directly related to the drag coefficient) to accommodate to the changes in speed of the bath. Clearly, if such changes occur within a macroscopic time t_m that is not large compared with τ, then the particle velocity might differ substantially from that of the gas. Thus particles in a suspension may precipitate at high speeds against solid objects placed in their way, allowing for a rather unusual and efficient transport process commonly called *inertial impaction*. Such a phenomenon can be quantified

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† The qualitative common feature is that the particle–fluid mass ratio m_p/m is a large parameter, but the quantitative value of this ratio is 176 for a H_2–UF_6 mixture and 10^{10} for a 1 μm water droplet in air.

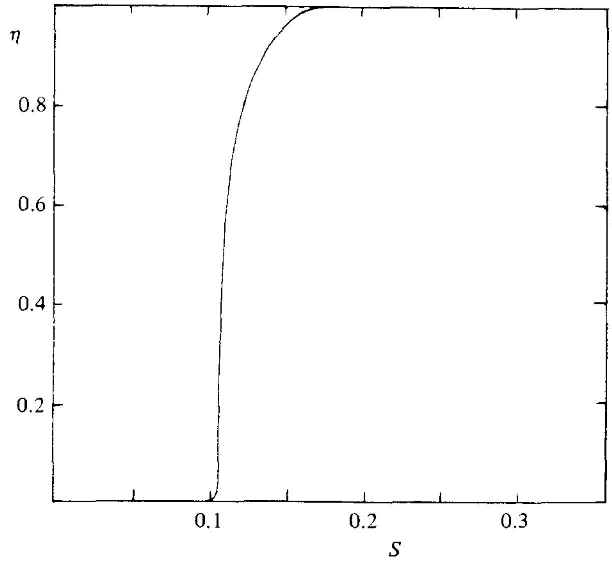

FIGURE 1. Typical response curve of an aerosol impactor. η is the fraction of the incoming aerosol that gets captured in the impaction plate at a particular value of the Stokes number S. Actual responses (experimental and calculated) of aerosol impactors can be found in Fuchs (1964), Marple & Willeke (1979) or Willeke & Pavlik (1978, 1979).

in terms of the so-called Stokes number S, or ratio between the microscopic and macroscopic times τ and t_m,

$$S \equiv \tau / t_m, \tag{1}$$

and has the property of entering into the picture in a most abrupt fashion. Its remarkable nonlinearity has been well documented theoretically and experimentally in the aerosol literature (Fuchs 1964; Friedlander 1977; Marple & Willeke 1979; Willeke & Pavlik 1978, 1979). Figure 1 shows a typical plot of the probability of particle capture (impact) η for a dispersion carried in a jet that impinges on a wall, as a function of the parameter S. For S sufficiently small, the aerosol is dragged along with the gas, skipping the obstacle. But above a threshold value S_* (the critical Stokes number) the capture efficiency rises quite abruptly to unity. Not surprisingly, the sharpness of that response has been exploited in practice to segregate particles of different sizes (masses) in instruments called impactors. As the fate of particles through impactors can be predicted without fundamental difficulties by treating them as deterministic objects obeying Newton's equations, the degree of perfection achieved by aerosol impactors has been quite considerable (Fuchs 1964; Friedlander 1977; Marple & Willeke 1979).

Given that background, we posed ourselves the question of whether or not it would be possible to obtain a response as abrupt as that of figure 1 when operating an impactor with heavy molecules instead of particles. If the answer were to be affirmative, then the consequences to the worlds of disparate-mass mixtures and ultrafine aerosols would be far reaching. For the aerosols, it would imply that one could in principle reduce the sizes of inertially capturable particles from the range 200–500 Å (Hering *et al.* 1979) currently available,† down to atomic dimensions. It

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† The impactor models Mark 5, 10 and 20 commercialized by Pollution Control Systems Corp, are supposedly able to capture particles down to 200 Å.

would also make it possible to use heavy molecules (stable, all identical and easy to generate and detect) as convenient laboratory models to experiment on the dynamics of fine mists (unstable, never monodisperse, and difficult and expensive to generate and detect). In the field of disparate-mass mixtures the implications would also be important. At the fundamental level, the response of molecular impactor would be governed by diffusion at the smaller, and by inertia at the larger, values of S. Its behaviour at intermediate values would thus display a most unusual transition between the purely stochastic and the purely deterministic, something worth studying as a special chapter in gas-phase transport processes. On the more practical side, one should note that much of what we know on disparate-mass mixtures was learned in the hope of achieving an efficient scheme to separate uranium isotopes through strong decelerations of UF_6–H_2 mixtures. Quite clearly, if a response anywhere near to that of figure 1 could be obtained in molecular impactors, their corresponding ability to separate isotopes would be far greater than that of any other available gasdynamic scheme. At this point we might be called to moderate our fantasy by a critical remark. If the proposed impaction scheme is so wonderfully promising, why has it not been tried yet? Wouldn't it be that the physical picture of a heavy atom moving deterministically through a light carrier gas is too far fetched to be of any use? Indeed, before taking the Reis–Fenn analogy as a reasonably well-founded conceptual scheme capable of guiding our thinking, one would have to make it compatible with the hard fact that the characteristic thermal velocity of an atom as heavy as mercury exceeds 100 m/s at room temperature. One would also be forced to demonstrate that the Stokes number of a heavy molecule can realistically reach values of order unity. Fortunately, however, the importance of the thermal speed has to be measured in relation to the mean streaming velocity. Accordingly, if our Hg atom is carried at 1000 m/s in a (still subsonic) hydrogen stream, its evolution might quite reasonably be considered as nearly deterministic. As a result, one could confidently go through the exercise of determining the drag coefficient of such a 'quasi-deterministic' atom moving through a light carrier gas, and establish that, indeed, its Stokes number might reach values of order unity in subsonic flows where the carrier fluid is still behaving as a continuum (Fernández de la Mora *et al.* 1981; Fernández de la Mora 1982). It is therefore quite clear that the reasons why molecular impactors have not been tested have little to do with the absurdity or the lack of promise of the scheme. Accordingly, the present paper will explore experimentally the phenomenon of heavy-molecule impaction. We shall first discuss the criteria needed to design a molecular impactor. Then we report on the actual performance of our first prototype, and compare it with that typical of aerosol impactors with the help of some theoretical considerations. The theory, however, is only valid in the region of small and large values of the Stokes number, where the kinetic problem is amenable to a hydrodynamic treatment. Finally, we conclude by putting our results in perspective of what is known in the fields of aerosol and heavy-molecule aerodynamic separation.

2. Design considerations and experimental apparatus

2.1. *Design*

The criteria to be followed in designing a molecular impactor do not differ in principle from those well known in the field of aerosols (Marple & Willeke 1979; Hering *et al.* 1979). Most importantly, the Stokes number of the heavy molecules has to reach values of order unity. For the purposes of design, we adopt the rough assumption that the hydrodynamic force acting on the heavy species as a result of its interaction with the carrier is linear in their relative velocity, with a proportionality coefficient (the friction factor) f related to the heavy–light diffusion coefficient D through Einstein's law

$$D = kT/f \tag{2}$$

(k is Boltzmann's constant and T the light-gas absolute temperature). Then the particle relaxation time τ ($= m_p/f$) can be written as

$$\tau = \frac{D\rho}{p}\frac{m_p}{m}, \tag{3}$$

where ρ, p and m are the light-gas density, pressure and molecular mass respectively, and m_p is the molecular mass of the heavy constituent. For a geometry in which a jet emerges from a nozzle with a speed U_n and encounters a solid wall downstream within a distance L of the order of the nozzle diameter d_n, the macroscopic time is $t_m = d_n/U_n$, so that the Stokes number becomes

$$S = \frac{D\rho}{p}\frac{m_p}{m}\frac{U_n}{d_n}. \tag{4}$$

It is worth noticing that the group $D\rho$ for heavy molecules is given by the Chapman–Enskog theory (Chapman & Cowling 1970), having a value comparable to the light-gas viscosity μ. We therefore introduce the diffusivity ratio

$$S_c = \mu/\rho D, \tag{5}$$

a quantity of order unity, weakly dependent on the temperature, and usually called the Schmidt number. If we also introduce the two variables determining the jet hydrodynamic properties, its Mach and Reynolds numbers

$$R \equiv \frac{\rho U_n d_n}{\mu}, \quad M \equiv \frac{U_n}{(\gamma p/\rho)^{\frac{1}{2}}} \tag*{(6), (7)}$$

(γ is the specific heat ratio for the light gas), then S can be written as

$$S = FM^2/R, \tag{8}$$

where the constant F (for Fenn) is a large number characteristic of the heavy–light mixture:

$$F = \gamma m_p/mS_c. \tag{9}$$

It is also clear that the group M^2/R is characteristic only of the light gas, representing the ratio of its own relaxation time μ/p to the hydrodynamic time t_m. Such a ratio is usually called a Knudsen number, and has to be small compared with unity in order for the gas to behave as a continuum. Accordingly, the group F (the Fenn number) represents a premium factor enabling S to reach values of order one while M^2/R is still small. F is a fair measure of the disparity of timescales characterizing our two-fluid system, and no inertial effects can be expected unless it takes substantial values, of the order of 100. Unfortunately for our purposes, it is not likely that we might find mixtures in which F is of the order of 10^3 while the heavy species has still a reasonably high volatility to form a stable suspension. Therefore the relative smallness of F sets narrow limits to the possibility of operation of molecular impactors. If S is to reach values of order one in a subsonic flow ($M < 1$), then the corresponding jet Reynolds number can be at most as large as F (equation (8)). Molecular impactors are thus called to operate at moderate values of the Reynolds number ($R \lesssim 100$) and possibly under compressible or even supersonic conditions. Since little is known on the performance of supersonic or compressible aerosol impactors, we will consider mainly subsonic situations. Low-Reynolds-number effects have been studied in some detail (Farthing 1983), but not when coupled with compressibility. The principal problem here is that the nozzle has to be sufficiently long to accelerate the heavy species efficiently up to the gas speed, at the cost of large pressure losses. Molecular impactors will thus have to tolerate a considerable velocity lag in the accelerating section in order to partially reduce the strong viscous effects through the nozzle.

2.2. *Control of the Stokes number*

For the purposes of this first experimental exploration, we chose R to be around 30. Since the parameter S had to be varied, we had to change either F or M. The former possibility is experimentally complicated, and has the additional inconvenience that the diffusivities for most of the heavy substances we had in mind are not known with sufficient accuracy (also see below). We thus decided to control S through M. Such a choice is clearly not beyond reproach, since the structure of the jet depends on M. It would not be clear then whether the observed result is a Stokes-number or a Mach-number effect. Fortunately, however, compressibility is not very important at values of M as large as 0.4 or even 0.5, so that not much M-effect ought to be expected in the response curve of an impactor to S below $M = 0.5$. Such an expectation is confirmed experimentally by the work of Hering *et al.* (1979) (the only available calibration for an impactor stage well into the compressible regime), where the standard subsonic calculation for the stage response agreed well with the measured behaviour when operating at $M \approx 0.6$. The role of compressibility in aerosol impaction has been considered also by Israel & Rosner (1983), who conclude that increasing the Mach number decreases the importance of inertia rather than increasing it. This implies that the response we might observe by controlling S through the Mach-number handle is actually less strong than in the absence of compressibility. A third control alternative would have been to vary R and keep M constant. But the Reynolds-number dependence of the jet structure at the small values at which we are forced to operate is surely far from negligible. Finally, the alternative of controlling S through F (by using different mixtures) encounters the new difficulty that the impactor performance depends not only on S, but also on the diffusivity ratio S_c, since diffusion is quite important here. One would then have to go through the taxing exercise of varying F by picking mixtures with different ratios m_p/m, but having the same value of the group S_c. As is often the case when making models, it is difficult to reproduce faithfully all the parameters of the real world.

2.3. *Apparatus*

Our design differs little from that of other low-pressure impactors discussed in the literature (see e.g. Hering *et al.* 1979). The main features of the apparatus are sketched in figure 2. The carrier gas (He) goes through a needle valve where the mass flow (and thus the Reynolds number) is controlled, passes through a critical orifice 0.2 mm in diameter and reaches the upstream chamber. There it flows over an electrically heated tungsten filament, which serves as a source of tungsten oxide molecules. The gas entrains evaporating heavy species, expands through a converging nozzle with exit diameter $d_n = 1.59$ mm, forming a jet that enters the downstream chamber with a speed U_n and finally impinges on a glass plate. The flow from the downstream chamber is removed by a mechanical pump at a nominal rate of 160 dm^3 per minute, through a throttling valve, which permits regulation of the jet Mach number M. The deter-

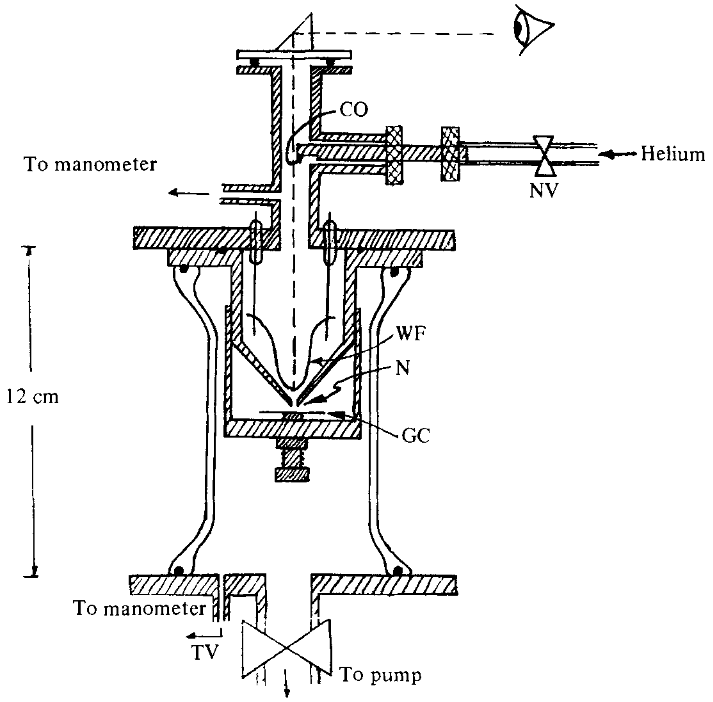

FIGURE 2. Sketch of the apparatus: NV, needle valve (control of R); CO, critical orifice (0.2 mm in diameter); WF, tungsten filament (0.25 mm diameter); N, nozzle (1.59 mm diameter); GC, glass cover slip; TV, throttling valve (control of M).

mination of M cannot be based on the assumption of isentropic behaviour commonly used in gasdynamics. At our modest values of R a significant portion of the pressure difference between the two chambers is lost through friction. Also, the velocity profile is far from flat, so that one has to use some average velocity to characterize U_n. For convenience, we based it on the mass flow $\dot{m}$, measured with a soap-bubble flowmeter:

$$\dot{m} \equiv \rho_n U_n A_n, \tag{10}$$

where $A_n = \frac{1}{4}\pi d_n^2$ is the cross-section at the nozzle exit (2 mm^2; N in figure 2). Right at the nozzle exit the pressure is equal to the background downstream pressure p_2, while the temperature can be calculated by assuming that the expansion is adiabatic (not isentropic), to yield

$$\frac{\dot{m} c_0}{\gamma p_2 A_n} = M[1+\tfrac{1}{2}(\gamma-1)M^2]^{\frac{1}{2}}, \tag{11}$$

where c_0 is the speed of sound for the carrier gas in the upstream (stagnation) chamber. M can then be determined by measuring $\dot{m}$ and p_2 (notice that (11) is quadratic in M^2). By measuring the pressure p_1 above the nozzle, we could judge on the self-consistency of the method by inferring an effective friction coefficient for the nozzle (Shapiro 1953), and checking its independence of M within the incompressible regime. The prediction of (11) was also checked favourably at the sonic point, which is easily determined experimentally as the curve $p_1(p_2)$ reaches a saturation. For the determination of the sound speed c_0 it was assumed that the gas stagnation temperature was not significantly altered by the hot filament. The validity of such an assumption was confirmed with a thermocouple and through the effect of lighting

FIGURE 3. Interference fringes formed by reflection of red light on a deposit of tungsten oxide.

up the filament on p_1 and p_2, both methods yielding temperature increments of the order of 5 °C. All pressures were measured with oil or mercury manometers.

The experimental constraint of keeping R constant while varying M is achieved automatically through the constancy of the mass flow $\dot{m} = \frac{1}{4}\pi\rho U_n d_n^2$, since from (6) and (10) $R = 4\dot{m}/\pi\mu d_n$. The hot-filament temperature is measured with an optical pyrometer, and is maintained at 1400 °C, where the evaporation of W atoms is negligible, but tungsten oxides (WO_x) vaporize readily. A small amount of air (1 % molar fraction, small enough to prevent nucleation) is allowed into the He, and some of its oxygen reacts with the filament, producing oxides,† which constitute the heavy molecules in these experiments. Nearly all the oxide is collected on the impaction plate, forming a transparent axially symmetric deposit whose shape can be inferred from the dark rings formed by interference of the light reflected at the WO_x–air and the WO_x–glass interfaces (figure 3). We obtain an accurate reading of the radial position of each ring using red light in a microscope with a micrometric movable crosshair. The shape of a few deposits (normalized to their maximum value) is shown in figure 4, where they can be seen to be linear in r^2 near the origin, with a slope changing greatly as a function of the Stokes number. As a convenient means to quantify the size of the deposit, we use the radial position r_0 at which its height is a factor e less than the maximum (i.e. $h(r_0) = h_{max}/e$). Unfortunately, the standard impactor η versus S curve cannot be used here because, unlike particles, WO_x molecules have a thermal speed of the order of 100 m/s and diffuse to the wall sooner

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† WO_2, WO_3 and W_2O_6, according to Schissel & Trulson (1965). The values of the Fenn number F for WO_3, W_2O_6 in He can be estimated from Tolfo (1975) to be 57 and 85 respectively.

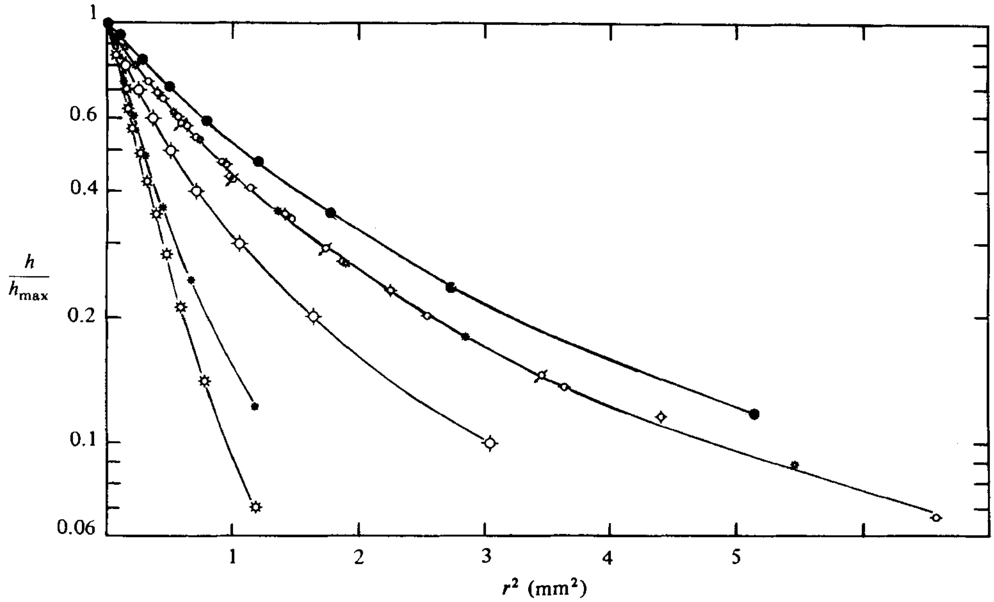

FIGURE 4. Normalized height distribution of various deposits. For $R = 36.9$: ●, $M^2 = 0.0106$; ○ ◇ ⌀ ✲, 0.0225; ◇, 0.049; ✡, 0.218. For $R = 16.5$: •, 0.035.

or later. Thus nearly all the incoming oxide is collected on the impaction plate ($\eta = 1$) at any value of S. To check that point we added a second impactor downstream from the first and observed that it never collected any appreciable deposit. Nonetheless, inertia leads to great variations in the width of the condensate (r_0^2). At small values of S, the WO_x molecules have a nearly nil mean normal speed close to the impaction plate, disposing of ample time to spread radially before condensing. But at larger values of S they retain their large initial velocity until hitting the wall, and have little time to disperse radially. Accordingly a curve $r_0^{-2}(S)$ would provide a quantitative measure of the effect of inertia. Such a curve is given in figure 5, showing again how strongly the deposition process is affected. The question now is to relate our experimental variable r_0^{-2} to the standard impactor capture efficiency η, before we can make any comparison between our results (figure 5) and those typical of particle impactors. Clearly, since η is an overall mass-transfer coefficient, its analogue in this problem would be a local mass-transfer coefficient: the ratio of the mass flux deposited on a given differential area to the free-stream mass flow heading towards that particular section (the so-called Stanton number St). The value St_0 of the Stanton number at the stagnation point will thus be a fair measure of the capture efficiency. But as the rate of deposition normalized by the free-stream flux is proportional to the deposit height divided by the deposit volume, and that quantity is proportional to the inverse deposit area, it follows that our ordinate in figure 5 is roughly proportional to St_0. The comparison between the two figures becomes now meaningful, since the local and the overall Stanton numbers for the capture of particles have nearly the same step-like response to the Stokes number. A more rigorous way to defend that point will be brought forth in the theoretical analysis below. Clearly, the steepness of our data and the facts that they saturate at values of S of order unity and that the mass-transfer coefficient is so much larger in the inertia-dominated regime than in the diffusion region make the two pictures very much alike. How-

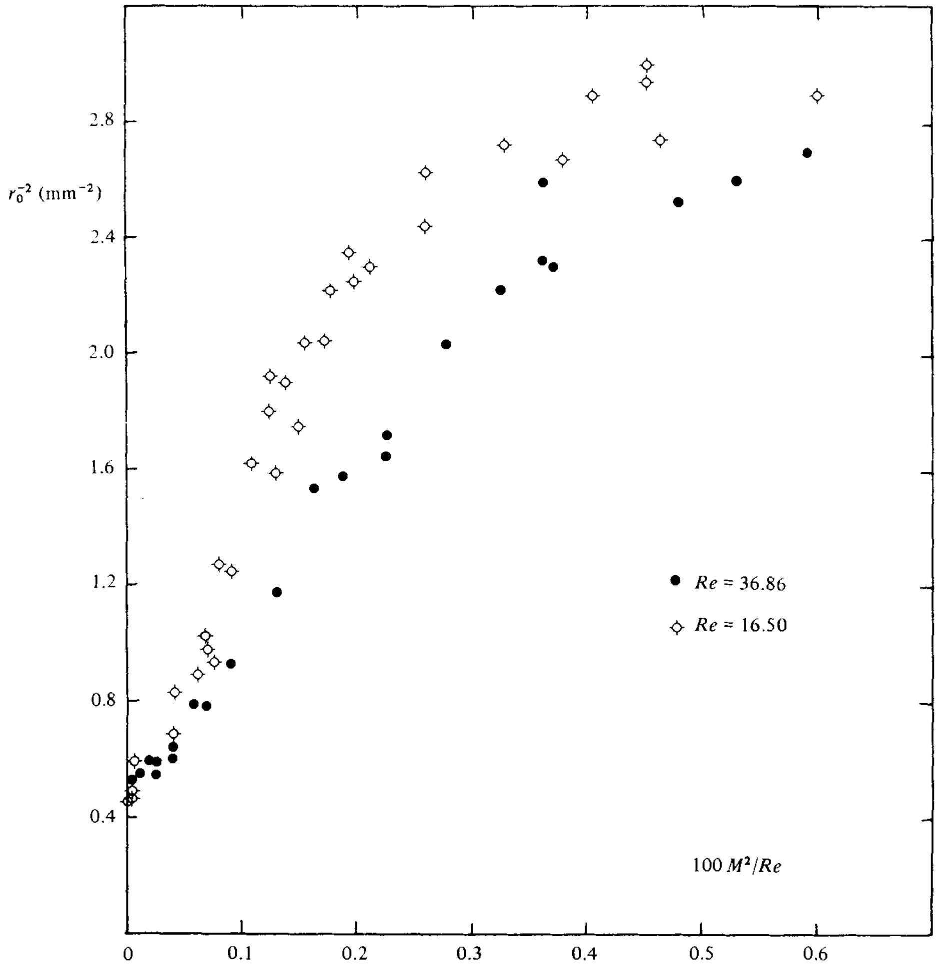

FIGURE 5. Inverse characteristic width r_0^{-2} of the WO_x deposits as a function of the inertia parameter. r_0^{-1} is obtained from curves analogous to those in figure 4 through the condition $h(r_0) = h_{max}/e$.

ever, the differences are also obvious enough. Most interestingly, no discontinuity reminiscent of the critical Stokes number of aerosol impactors is visible. The observation might seem surprising, because one can predict a change in nature on the system's response at a critical rate of deceleration, even when the particles have a non-negligible diffusivity (Fernández de la Mora 1982). Yet there is really no such discontinuity in figure 1, but rather a dependence similar to that of the function $\exp(-1/(S-S_*))$. The sharpness of the response is due mainly to the fact that η is absolutely zero at first, and then increases to values of order unity within a small interval of the Stokes number. For the molecular impactor, however, since the diffusive rate of capture is less than an order of magnitude smaller than the maximum, the capture efficiency curve can only rise over that modest order of magnitude. Such a result could have been easily predicted, since the ratio $\eta(\infty)/\eta(0)$ between the maximum and the minimum values of η is nothing but the inverse of the mass-transfer Stanton number, proportional to the $\frac{1}{2}$ power of R and the $\frac{2}{3}$ power of S_c. The group takes enormous values for aerosols, because R can be quite large, and especially because S_c generally exceeds 10^4. But for heavy molecules S_c is of order unity, and R is bound by the modest available values of F. The effect of diffusion at subcritical values of S is therefore of sufficient importance to overshadow such an interesting effect as the subcritical response of particle impactors.

3. Theoretical considerations

In this section we bring forth some theoretical information to help interpret and rationalize our experimental results. To proceed with rigour, one would first have to determine the carrier-fluid velocity field, a non-trivial task given the fact that none of the standard convenient limits $M \rightarrow 0$ or $R \rightarrow \infty$ is strictly applicable. Nonetheless, the $M \rightarrow 0$ limit could in principle be used to provide the fluid-velocity field for most of the conditions investigated in figure 5. Next one would have to find the fate of the heavy species in order to predict the deposit shape. The continuum (diffusion) equations would provide a fair description of the behaviour, but only in the region where $S \lesssim 0.04$ (Fernández de la Mora 1982). Even so, most of the progress would also have to be numerical. In the region where $S \gtrsim 1$, the heavy-species velocity remains quite high down to the point of impact. Its thermal speed could thus be neglected throughout (a hypersonic theory), allowing for a deterministic treatment exactly as one does with aerosols. But at intermediate values of S neither the hypersonic nor the hydrodynamic approach would work, since S is large enough to drive the system far from equilibrium, yet the impact speeds are not sufficiently high to make the thermal agitation negligible. The problem thus calls for a kinetic treatment. Fortunately, the large mass disparity and the diluteness of the heavy component permit an extreme reduction of the heavy-species Boltzmann equation to a linear partial differential equation of the Fokker–Planck type (Fernández de la Mora & Mercer 1982). Still, that kinetic equation has four independent variables (two in velocity space, two in real space) for an axisymmetric geometry, so that the problem still remains too complex even for numerical treatment. Accordingly, we shall have little to say on the nearly intractable fully kinetic, or on the well-known hypersonic (deterministic) regions, and will concentrate on the diffusion domain where $S \ll 1$. In that limit, the continuum analogue of inertia is pressure diffusion, as shown by Ramshaw (1979, 1981) and Fernández de la Mora & Rosner (1982). The purpose of the following analysis is to obtain an expression for the quantity r_0^{-2} previously obtained experimentally, which is approximately related to the curvature at the origin ($r = 0$) of the rate of deposition $j''(r)$:

$$r_0^{-2} = \left\{ \frac{\mathrm{d}^2[j''/j''(0)]}{\mathrm{d}r^2} \right\}_{r=0}. \tag{12}$$

Its determination requires only a relatively simple local analysis near the axis of symmetry, and can be carried out analytically to lowest order in the small parameter $(RS_c)^{-1}$, the inverse Péclet number. The steady-state mass-conservation equation for the heavy species is

$$\nabla \cdot (\rho_p \boldsymbol{u}_p) = 0, \tag{13}$$

where ρ_p is the density field and $\boldsymbol{u}_p$ the velocity field for the heavy species. $\boldsymbol{u}_p$ can be expressed as†

$$\boldsymbol{u}_p = \boldsymbol{u} - D \nabla \ln \rho_p - \tau (\boldsymbol{u} \cdot \nabla) \boldsymbol{u} + O(S^2) \tag{14}$$

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† Equation (14) states the balance of the drag forces $(\boldsymbol{u}_p - \boldsymbol{u})/\tau$ with the partial pressure (diffusion term) and the particle acceleration (inertial term), equal to lowest order to the fluid acceleration $(\boldsymbol{u} \cdot \nabla) \boldsymbol{u}$.

with errors of the order of S^2 (Fernández de la Mora & Rosner 1982), where $\boldsymbol{u}$ is the velocity field for the light gas. This result is similar to that given by the Chapman–Enskog (CE) theory in the dilute, high-mass-ratio limit, if one neglects thermal diffusion. However, the inertial term $\tau\boldsymbol{u}\cdot\nabla\boldsymbol{u}$ appears in (14) instead of the CE pressure diffusion. After making use of the momentum-conservation equation for the carrier (which relates $\boldsymbol{u}\cdot\nabla\boldsymbol{u}$ to the divergence of the pressure tensor), it appears that the principal difference between (14) and the CE theory is that (14) includes also the irreversible viscous stress. But the difference is a higher-order term in the CE expansion, so that (14) is not inconsistent with the standard first-order results of the kinetic theory of gases. We will therefore follow (14) for this calculation.

Let us consider the behaviour near the axis of symmetry of an incompressible fluid with a velocity given by

$$\boldsymbol{u} = -u(z)\,\boldsymbol{e}_z + \tfrac{1}{2}u'(z)\,r\boldsymbol{e}_r \tag{15}$$

in polar coordinates, where $u'(z) \equiv \mathrm{d}u(z)/\mathrm{d}z$, z is the distance to the wall along the axis of symmetry, and r is the radial (polar) coordinate. $u(z)$ is an unspecified function of z, which vanishes as z^2 near the wall ($z = 0$). The inertial velocity drift is then given by

$$\tau\boldsymbol{u}\cdot\nabla\boldsymbol{u} = \tau[uu'\boldsymbol{e}_z + \tfrac{1}{4}(u'^2 - 2uu'')\,r\boldsymbol{e}_r], \tag{16}$$

and the mass-conservation equation becomes

$$-(u+\tau uu')\frac{\partial\rho_{\mathrm{p}}}{\partial z} + \tfrac{1}{2}r[u' + \tau(uu'' - \tfrac{1}{2}u'^2)]\frac{\partial\rho_{\mathrm{p}}}{\partial r} - D\,\nabla^2\rho_{\mathrm{p}} = \tfrac{3}{2}\tau u'^2\rho_{\mathrm{p}}. \tag{17}$$

We shall base our method of solution in the smallness of the diffusion term $D\,\nabla^2\rho_{\mathrm{p}}$, following the method of subcharacteristics (Cole 1968). Making (17) non-dimensional by use of the characteristic length d_{n} and speed U_{n}, it becomes clear that the diffusion term is small, of order $(S_{\mathrm{c}}\,R)^{-1}$ ($\sim 10^{-2}$). Dropping it, (17) becomes a first-order partial differential equation, which can be solved following the characteristics (trajectories) along which

$$\frac{-\mathrm{d}z}{u+\tau uu'} = \frac{2\mathrm{d}r/r}{u' + \tau(uu'' - \frac{1}{2}u'^2)} = \frac{\mathrm{d}\rho_{\mathrm{p}}/\rho_{\mathrm{p}}}{\frac{3}{2}\tau u'^2}. \tag{18}$$

Defining

$$\ln G(z) \equiv -\frac{3\tau}{2}\int_0^z \frac{u'^2}{u+\tau uu'}\,\mathrm{d}z, \tag{19}$$

each trajectory can be traced by assigning a constant value of ξ in the equation

$$\xi \equiv rG(z)\,(u+\tau uu')^{\frac{1}{2}}, \tag{20}$$

and along trajectories ρ_{p} is given by

$$\rho_{\mathrm{p}} = NG(z), \tag{21}$$

where $N = N(\xi)$ is constant along trajectories. Introducing now the natural dependent variable N and independent variables ξ, r, and neglecting diffusion along the trajectories, there results

$$G^{-2}(z)\,\partial_z N + D\xi^{-1}\,\partial_\xi(\xi\,\partial_\xi N) = 0. \tag{22}$$

For the case where the heavy species is introduced at the point $r = 0$, $z = z_0$ with no radial spread, the solution is

$$N = Cs^{-1}\exp\left[-\frac{\xi^2}{4Ds}\right], \tag{23}$$

where C is a constant related to the amount of material injected at $z = z_0$, and s is a z-like variable defined by

$$\mathrm{d}s = -G^2(z)\,\mathrm{d}z \tag{24}$$

and an appropriate initial condition (see below). Determining the deposition rate at the wall now requires considering diffusion in the z-direction across a thin boundary layer near $z = 0$. The analysis cannot be completed exactly for the case when $N(\xi)$ away from the wall is not a constant. But to obtain the magnitude r_0^{-2} it suffices to expand $N(r, z)$ and $\boldsymbol{u}(r, z)$ in powers of r, retaining only terms up to r^2. In that way, we will reduce our partial differential equation to two ordinary differential ones. Exploiting the existing symmetries, we may write

$$\rho_\mathrm{p} = \rho_0(z) + r^2\rho_1(z) + \ldots, \tag{25}$$

$$u = -\boldsymbol{e}_z z^2(\alpha_0 + \alpha_1 r^2 + \ldots) + \boldsymbol{e}_r zr(\alpha_0 + \tfrac{1}{2}\alpha_1 r^2 + \ldots), \tag{26}$$

where the constants α_0 and α_1 are functions of R given by solving the hydrodynamic problem for $\boldsymbol{u}$. Substituting (25) into (17) and introducing the inner variable

$$\eta \equiv z(\alpha_0/D)^{\frac{1}{3}}, \tag{27}$$

there results

$$\rho_0'' + \eta^2\rho_0' = 0, \tag{28}$$

$$\rho_1'' + \eta^2\rho_1' - 2\eta\rho_1 = -\eta^2\alpha_1\alpha_0^{-1}\rho_0', \tag{29}$$

where terms of order $(D/\alpha_0)^{\frac{1}{3}}$ or smaller have been neglected, and primes denote differentiation with respect of η. The boundary conditions are

$$\rho_0 = \rho_1 = 0 \quad \text{at} \quad \eta \to 0, \tag{30}$$

while, as $\eta \to \infty$, ρ_0 and ρ_1 have to match with the outer solution given by (23). Accordingly, defining

$$\rho_\mathrm{w} \equiv CG(0)/s(0), \tag{31}$$

the boundary (matching) conditions as $\eta \to \infty$ are, from (23),

$$\rho_0 \to \rho_\mathrm{w}, \tag{32}$$

$$\rho_1 \to \frac{-\rho_\mathrm{w}\,G^2(0)\,\alpha_0\,z^2}{4Ds(0)}. \tag{33}$$

The solution for ρ_0 is

$$\rho_0(\eta) = \frac{\rho_\mathrm{w}\displaystyle\int_0^\eta \exp(-\tfrac{1}{3}x^3)\,\mathrm{d}x}{3^{\frac{1}{3}}\Gamma(\frac{4}{3})}, \tag{34}$$

while ρ_1 can be written by superposition as

$$\rho_1 = -\left(\frac{\alpha_0}{D}\right)^{\frac{1}{3}}\frac{\rho_\mathrm{w}}{4s(0)}g(\eta) - \frac{\alpha_1}{\alpha_0}\frac{H(\eta)\,\rho_\mathrm{w}}{3^{\frac{1}{3}}\Gamma(\frac{4}{3})}, \tag{35}$$

where

$$H'' + \eta^2 H' - 2\eta H = \eta^2\exp(-\tfrac{1}{3}\eta^3), \quad H(0) = H(\infty) = 0, \tag{36}$$

and

$$g'' + \eta^2 g' - 2\eta g = 0, \quad g(0) = 0, \quad g(\infty) \to \eta^2. \tag{37}$$

Both g and H are easily found in a Taylor expansion with infinite radius of convergence. The magnitudes of interest giving the rate of deposition are $g'(0)$ and $H'(0)$, which we obtain as

$$g'(0) = 1.445, \quad H'(0) = -0.199. \tag{38),(39}$$

The rate of deposition is proportional to $(\partial N/\partial \eta)_{\eta=0}$:

$$\left(\frac{\partial N}{\partial \eta}\right)_{\eta=0} = \rho_0'(0) + r^2 \rho_1'(0) + \dots, \tag{40}$$

whence it results from (12) that

$$r_0^{-2} = H'(0)\frac{\alpha_1}{\alpha_0} + \frac{g'(0)\, 3^{\frac{1}{3}} \Gamma(\frac{4}{3})\, (\alpha_0/D)^{\frac{1}{3}}}{4s(0)}. \tag{41}$$

All of the dependence on the Stokes number is contained in $s(0)$, given by integration of (24). The usual initial condition for (24) is that $s(z_0) = 0$. For our experiments, however, the point of release of the heavy molecules ($z = z_0$) was a few nozzle diameters upstream from the throat, in a region sufficiently wide for the group RS_c not to be a large parameter. Accordingly, there is ample time for diffusion to fill most of the space with the heavy species. Nonetheless, our treatment is valid in the region downstream from the nozzle, so that we can impose the boundary condition for (24) at the nozzle throat ($z = L$). An appropriate parameter to fix there is the width of distribution N:

$$r_\infty^2 \equiv \frac{4Ds(L)}{G^2(L)\, U_\infty}, \tag{42}$$

where we have defined

$$U_\infty \equiv u(L). \tag{43}$$

Such a choice determines $s(0)$ as

$$s(0) = \frac{G^2(L)\, U_\infty}{4D} r_\infty^2 + \int_0^L G^2(z)\, \mathrm{d}z. \tag{44}$$

Now, because diffusion is so efficient in the slow region between the filament and the nozzle, most of the space is seeded with the heavy component, and r_∞ is of the order of the nozzle diameter d_n. But $G^2(z)$ is unity except for small corrections of order S, so that the second term in (44) is smaller than the first $[s(L)]$ by a factor $S_c R$, and can be neglected. Furthermore, most of the dependence of $s(0)$ on S is contained in the term $G^2(L)$, which incorporates the strong accelerations occurring between the nozzle and the plate. Clearly, the acceleration above $z = L$ has to be much milder than below $z = L$, because otherwise the same inertial effects occurring in the deceleration region would occur in the accelerating one. Accordingly, the local Stokes number $\tau u'$ has to be much smaller in the region upstream than in the region downstream from the nozzle if the light gas is to be able to accelerate the heavy molecules. r_∞ is thus nearly independent of S (though it could perhaps depend on R). Actually, r_∞ plays an interesting role in this problem, because in the inertia-dominated region there is no chance to change the width of $\rho_p(r)$ between the nozzle and the wall, so that

$$r_\infty = \lim_{S \gg 1} r_0(S). \tag{45}$$

Notice finally that our expansion is valid only for small values of r/d_n. However, the corresponding prediction for the curvature at the origin r_0^{-2} (12) does not depend on whether the resulting group $(d_n/r_0)^2$ is large or small.

3.1. *High-Reynolds-number behaviour*

Obtaining a numerical prediction for $r_0^{-2}(S)$ requires a previous complete specification of the fluid velocity field $\boldsymbol{u}(r, z)$, which one would have to obtain for every particular value of R and every geometry. Therefore it is instructive to examine the actual dependences of the parameters α_0, α_1 and $G^2(L)$ on R and S in the limit of large Reynolds numbers, keeping in mind that the predictions that follow contain substantial errors, of order $R^{-\frac{1}{2}}$. We assume that the velocity field is given by (Schlichting 1968)

$$\boldsymbol{u} = -\boldsymbol{e}_z 2(a\nu)^{\frac{1}{2}}\phi(\xi) + \boldsymbol{e}_r ra\phi'(\xi), \tag{46}$$

with

$$\xi \equiv (a/\nu)^{\frac{1}{2}} z, \tag{47}$$

while a is the velocity gradient

$$a \equiv U_\infty / L, \tag{48}$$

ν is the light-gas kinematic viscosity

$$\nu \equiv \mu/\rho, \tag{49}$$

and $\phi(\xi)$ is a known function. Then

$$\ln G^2(L) = -6a\tau \int_0^{L(a/\nu)^{\frac{1}{2}}} \frac{\mathrm{d}\xi\, \phi'^2}{\phi} \tag{50}$$

has the asymptotic value

$$\ln G^2(L) = -6a\tau \left\{ 2.167 + \ln\left[L\left(\frac{a}{\nu}\right)^{\frac{1}{2}} - 0.569 \right] \right\}. \tag{51}$$

For our particular geometry where $d_{\mathrm{n}} = L$, the groups $a\tau$ and $L(a/\nu)^{\frac{1}{2}}$ are S and $R^{\frac{1}{2}}$ respectively. The constant α_0 follows from the behaviour of $\phi(\xi)$ near $\xi = 0$ $(\phi''(0) = 1.312)$:

$$\alpha_0 = 1.312(a^3/\nu)^{\frac{1}{2}}. \tag{52}$$

Finally, the constant α_1 cannot be given from this boundary-layer analysis, but depends on the structure of the impinging jet. Nonetheless, we can expect that the inviscid radial velocity field will have the form

$$u_r = ar\left(1 + \frac{\beta r^2}{d_{\mathrm{n}}^2} + \dots\right), \tag{53}$$

where the constant β is of order unity, in which case (Schlichting 1968, p. 227)

$$\frac{\alpha_1}{\alpha_0} = 1.1291\,\frac{\beta}{d_{\mathrm{n}}^2}. \tag{54}$$

Substituting all that information into (41), we obtain

$$\left.\begin{aligned} \left(\frac{r_\infty}{r_0}\right)^2 &= -0.225\beta\left(\frac{r_\infty}{d_{\mathrm{n}}}\right)^2 \\ &\quad + 2.0374 S_{\mathrm{c}}^{-\frac{2}{3}} R^{-\frac{1}{2}}\{2 + 6S[2.167 + \ln(R^{\frac{1}{2}} - 0.569)]\} \quad \text{for} \quad S \ll 1, \\ \left(\frac{r_\infty}{r_0}\right)^2 &\to 1 \qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad \text{for} \quad S \gtrsim 1. \end{aligned}\right\} \tag{55}$$

Accordingly, except for the presence of the term proportional to β, r_0^{-2} behaves as implied in our rough argument at the end of §2. An interesting consequence follows from the fact that the β-term can be controlled by changing the curvature of the obstacle, and could perhaps be such that $r_\infty / r_0(0) \ll 1$. In that case the sharpness of the response in figure 5 would be greatly increased.

4. Discussion

As mentioned previously, much of our knowledge on the peculiar dynamics of disparate-mass gas mixtures has been closely associated with technological efforts to achieve an efficient aerodynamic method of separating isotopes. The literature on the subject is considerable, and we shall not attempt a review here.† Instead, we will refer only to the 'separation nozzles' developed by Becker (1978) and his group at Karlsruhe. In these schemes, a mixture with a few percent molar fraction of a heavy species (UF_6) carried in a light gas (H_2) is accelerated to near-sonic speeds at Reynolds numbers of the order of 100. Subsequently the flow is forced to decelerate rather sharply, upon which the various isotopes follow different paths. The fluid is then divided into two streams, one of which is slightly enriched in the lighter isotope. In the best-studied process, the deceleration occurs centrifugally in a curved channel (Becker 1978). In another and rather interesting scheme (Bley & Ehrfeld 1981), the deceleration occurs frontally, through the impact of two identical opposed jets, both carrying UF_6. The region near the collision plane is then also enriched in the heavy isotope. The procedure is thus in many ways analogous to that of particle impactors. However, the conceptual schemes that guided progress in the two fields were quite different, in spite of the pioneering ideas of Reis & Fenn (1963). The lack of cross-fertilization is obvious from the technical features of the 'separation nozzles'. Although it is known among aerosol physicists that a frontal deceleration leads to a much sharper mass separation than a centrifugal deceleration (Willeke 1980), the version of the Karlsruhe nozzles that has reached the industrial stage is based on centrifugation (Becker *et al.* 1982). Interestingly enough, the opposing jet scheme studied later by Bley & Ehrfeld (1981) came geometrically very close to the opposed-jet aerosol impactor successfully developed by Willeke & Pavlik (1978, 1979). But it chose to impact two identical seeded jets, instead of a seeded jet against a clean one as in the aerosol tradition. It is therefore clear that the evolution of the 'separation nozzles' owes little to aerosol physics. On the other hand, although our molecular impactor was inspired by the Reis–Fenn analogy, its present capacity to separate isotopes leaves much to be desired. An appropriate physical (not economical) figure of merit measuring such separating powers is the logarithmic slope Π of the curve giving the flux of one species $j''(m_p)$ as a function of its mass m_p;

$$\Pi = \frac{\mathrm{d}\ln j''}{\mathrm{d}\ln m_p}. \tag{56}$$

This number is equal to $\frac{1}{2}$ for the traditional diffusion-separation process. Its value for the Karlsruhe nozzles is around two for the commercial process, but can be as high as four. For our molecular impactor no isotopic yields were measured, but the value of Π can be inferred from our figure 5 as

$$\Pi = \frac{\mathrm{d}\ln r_0^{-2}}{\mathrm{d}\ln S}, \tag{57}$$

since $\mathrm{d}\ln S/\mathrm{d}\ln m_p = 1$ and j'' is roughly proportional to r_0^{-2}. But for our experiments Π is below unity. In more recent measurements performed in our laboratory by Dr Akiko Natsume, better results for Π were achieved by varying the geometrical conditions. However, they never exceeded the value $\Pi = 2$. Our work thus fails to prove any significant superiority for our design based on the inertial conceptual

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† Some of the pertinent bibliography is discussed in Fernández de la Mora (1984).

scheme over others when it comes to separating isotopes. But we are still far from having reached an optimal configuration, while the mixed nature of our heavy species (WO_2, WO_3, W_2O_6) necessarily reduces the sharpness of the impactor's response.

Another important matter deserving discussion is whether or not one can definitely assert that inertia is the principal mechanism leading to the behaviour observed in figure 5. Among others, the Karlsruhe group that has led the field for over two decades has defended the alternative view that the cause of separation is pressure diffusion (Becker *et al.* 1977). We contend, however, that the concept of pressure diffusion (or any other continuum concept following from the Chapman–Enskog theory) is valid only when the appropriate Knudsen number (S here) is small. For small values of S inertia and pressure diffusion are roughly the same thing (Ramshaw 1979; Fernández de la Mora & Rosner 1982), and r_0^{-2} is linear in S. At higher values of S nonlinear effects ought to be expected, and can indeed be observed in figure 5. They first manifest themselves with a positive and then a negative curvature leading to a saturation tendency at large S. None of these three nonlinear features has much to do with pressure diffusion, all three being typical of an impactor's response to inertia (figure 1). The concept of heavy-molecule inertia is therefore more general than the near-equilibrium transport mechanism of pressure diffusion. Inertia is the natural generalization of the latter concept to describe the non-equilibrium flow conditions at which the aerodynamic separation process is most efficient.

5. Conclusions

This paper has explored the effects of heavy-molecule inertia on the transport of condensible tungsten oxide vapours carried by a subsonic helium jet and impacting on a solid surface. The shape of the deposits formed was seen to depend enormously on the Stokes number S, which was controlled while maintaining the jet structure practically unchanged. A local analysis in the vicinity of the stagnation point has been followed to demonstrate the linearity of the impactor's response to S in the near-equilibrium region $S \ll 1$. In this domain pressure diffusion and inertia are similar concepts. But the impactor's response at higher values of S is clearly nonlinear, with first a positive and then a negative curvature followed by a tendency towards saturation as $S \gtrsim 1$. Such a behaviour is similar to that of aerosol impactors, and cannot be explained without invoking the far from equilibrium effects due to the large inertia of the heavy species. The local rate of deposition at the stagnation point is seen to change by nearly an order of magnitude in the interval $0 \lesssim S \lesssim 1$. The response achieved thus far is not sharp enough to establish molecular impaction as a highly promising method for the aerodynamic separation of isotopes. Nonetheless, much room is left here for technical improvement. The present work documents for the first time the transition from the purely stochastic (diffusive) to the purely deterministic (inertial) mode of transport of a heavy condensable vapour.

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