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DENSITY AND SHAPE FACTOR
OF SODIUM AEROSOL

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W. Hinds and M.W. First

Harvard University
School of Public Health
665 Huntington Ave.
Boston, MA 02115

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ABSTRACT

This report covers the period January 1, 1976 to March 31, 1976. Several approaches for characterizing the physical and aerodynamic properties of irregular shaped aerosol particles are reviewed. Measurements of density modification factor, dynamic shape factor, and particle density using an aerosol centrifuge and a scanning electron microscope are described. Calibration procedures for this characterization method are described and preliminary results reported.

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INTRODUCTION

Progress during the second quarter (Jan. 1 - March 31, 1976) of project E(11-1)-2803 has been made in the development of techniques and calibration of equipment for the measurement of airborne density and shape of sodium aerosol particulate. The sodium aerosol was prepared by burning one pound of metallic sodium in a heated pot in a 90 m³ chamber. The aerosol generated was sampled with an aerosol centrifuge¹ which fractionates the particles according to aerodynamic size. Physical measurements were made on these fractionated particles by SEM (Scanning Electron Microscopy). These data permit the determination of the density modification factor (α) which allows Stokes's law to be used with the handbook density for the aerosol material. Measurements of mass and number concentration permit determination of airborne particle density and dynamic shape factor for nonspherical particles. Aerodynamic size calibration of the aerosol centrifuge was performed using particles of known size and density. Replicas of diffraction gratings were used to calibrate SEM photographs. A description of the sampling and analysis procedures and the preliminary results are given below.

THEORETICAL DESCRIPTION

Following a sodium fire accident, sodium fume would fill the LMFBR containment vessel and leak to the external environment at the rate of 0.1% of the containment volume per 24 hours. Although leakage rate will be constant, leakage of sodium fume will decrease with time principally because natural sedimentation will reduce airborne sodium concentration within the containment. To predict this effect with reasonable confidence, it is necessary to calculate the terminal settling velocity, V_{ts} , of the sodium aerosol particles accurately. This is done by combining Stokes's law, which applies only to uniform density spheres, with appropriate correction factors to account for (1) the nonspherical particle shape of the agglomerated sodium fume particles and (2) the non-handbook density of these particles.

Stokes's law applies to the steady state velocity situation where aerodynamic drag force equals gravitational force. Generally, a correction factor must be applied to both these forces to relate them to some measured or known dimensions of an irregular particle. A dynamic shape factor, χ , relates drag force on an irregular particle to that on a sphere of equal volume² and a volume shape factor, α_v , relates the actual volume of the irregular particle to some characteristic dimension cubed.³ The characteristic dimension is usually one that can be easily measured such as projected area diameter, D_{pa} , or Martin's diameter determined microscopically. These shape factors can also be defined as the proportionality constant between two characteristic diameters. The dynamic shape factor is the square of the ratio of the diameter of the equivalent

volume sphere, D_e , to the Stokes diameter of an irregular particle, the latter being D_e the diameter of a spherical particle with the same V_{ts} and density as the irregular particle. Similarly, the volume shape factor for the projected diameter is $\pi/6$ times the cube of the ratio of D_e to D_{pa} . An additional correction factor, the slip correction factor, C_c , is important when particle size approaches the mean free path of the gas.

The dynamic shape factor, χ , is usually determined by similarity experiments that measure V_{ts} of particles of known geometry and density in liquids. For these experiments there is no need for a volume shape factor, α_v , because the true value is known a priori. In aerosol studies with irregular particles, the volume is not known and cannot be measured directly. Consequently, it is necessary to calculate particle volume by using a volume shape factor and a dimension that can be measured, such as the projected area diameter, D_{pa} . Usually, α_v factors have been measured empirically for a specific process producing crushed materials having a known density (such as crushed quartz) and as such have limited general application. It is commonly assumed that α_v is constant with particle size for crushed materials. It is unreasonable to assume that α_v will be constant for agglomerated materials such as sodium fume.

The approach that has been used up to now for computer modeling of sodium fires^{4,5} was to calculate the equivalent volume diameter from the source term and coagulation theory and to use a combined correction factor, α , the density modification factor, which includes both an aerodynamic correction factor and a correction for unknown particle density. When properly used, this approach provides sufficient information to calculate V_{ts} , but does not provide information on the physical size of the particles which is important for thermal and electrostatic deposition and adsorption. Furthermore there is no theoretical basis for assuming a particular α value. The values that have been used are those that make the computer simulation agree with experimental data. Unfortunately, there is uncertainty as to the value of airborne particle density, ρ_p , of the sodium fume particles. This arises from (1) uncertainty of chemical composition because there will be an unknown mixture of oxide, peroxide, hydroxide, and carbonate, depending on CO_2 and H_2O concentrations, (2) an unknown amount of hydration of the sodium carbonate that is formed, (3) unknown porosity, and (4) variable amounts of interior interstices of agglomerated particles depending on how loose or compact the primary particles are clustered.

The density modification factor has been used in Stokes's law in the following manner:

$$V_{ts} = \frac{\alpha \rho_b D_e^2 C_c (D_e) g}{18 \eta} \quad (1)$$

where ρ_b is the handbook density for the aerosol material; g , the acceleration of gravity; $C_c (D_e)$, the slip correction for an equivalent volume sphere; and η is gas viscosity. Stokes's law can

also be expressed in terms of the aerodynamic diameter, D_a , i.e., the diameter of a unit density sphere that has the same settling velocity as the particle:

$$V_{ts} = \frac{D_a^2 C_c(D_a) g}{18 \eta} \quad (2)$$

and in terms of the equivalent diameter, D_e , a dynamic shape factor, χ , and the true particle density, ρ_p :

$$V_{ts} = \frac{\rho_p D_e^2 C_c(D_e) g}{18 \eta \chi} \quad (3)$$

From equations (1) and (2), the density modification factor, α , can be expressed as:

$$\alpha = \frac{D_a^2 C_c(D_a)}{\rho_b D_e^2 C_c(D_e)} \quad (4)$$

and from equations (1) and (3) as,

$$\alpha = \frac{\rho_p}{\rho_b \chi} \quad (5)$$

Assuming true particle density is known, there are three approaches to characterizing irregular particles with a device that fractions particles by aerodynamic diameter. One is to microscopically measure the projected area diameter of particles of known aerodynamic diameter. The ratio, $D_{pa}^2 \rho_p / D_a^2$, neglecting slip correction, is a combined shape factor which relates the microscopically measured diameter to the observed settling velocity. Whereas this ratio is relatively simple to determine, it cannot be used with diameters measured in other ways and provides little information on physical properties of the particles. A second approach, really a refinement of the first, is to split, α , the shape factor into the aerodynamic component related to the equivalent volume sphere, and the volume shape factor relating D_{pa} to D_e . Although this provides the desired information, it is now necessary to solve for two coefficients and it requires additional information which generally will be available only when particle density is known. The third approach involves direct estimation of the diameter of an equivalent volume sphere, D_e , and calculation of the dynamic shape factor from the ratio $D_e^2 \rho_p / D_a^2$. This approach provides the desired information but direct measurement of equivalent volume or volume diameter is difficult. One approach is to measure particle volume directly in a Coulter counter. The problem here is that the particles have to be fractionated according to aerodynamic diameter, and then suspended in a liquid for measurement.

Another approach is to measure D_{pa} microscopically and assume it is equal to D_e . This involves a significant error when particles are not approximately isometric. Finally, the equivalent diameter can be estimated directly from SEM photographs. This is a three dimensional analogue of measuring D_{pa} in two dimensions with a Porton graticule. It is possible to do this with SEM micrographs because they are photographs of three dimensional images rather than silouettes and can be compared visually to spheres of known volume.

When particle density is unknown it can be calculated from D_e and the average particle mass which is determined by measuring total mass, M , and count N , for a group of particles. If particle density is a function of particle size this calculation must be done for particles of known aerodynamic diameter. The equation for particle density is:

$$\rho_p = \frac{6M}{\pi \cdot D_e^3} N \quad (6)$$

and the dynamic shape factor is,

$$\chi = \frac{\rho_p D_e^2 C_c(D_e)}{D_a C_c(D_a)} \quad (7)$$

where $C_c(D_e)$ and $C_c(D_a)$ are slip correction factors for the respective diameters.

EXPERIMENTAL PROCEDURE

Sodium fume has been prepared by heating to combustion a one³ pound charge of metallic sodium and allowing it to burn in a 90 m³ chamber. The aerosol is sampled with an aerosol centrifuge¹ for 10 to 50 minutes depending on chamber concentration. The sample inlet is located 2 meters above the aerosol chamber floor. The centrifuge, shown in figure 1, acts like a spectrometer and continuously fractionates the aerosol according to aerodynamic diameter along a removable 40 cm metal foil. Every point along the foil is characterized by a known aerodynamic diameter. At 1100 rpm, 6 lpm of the chamber atmosphere enters the centrifuge and 75 ml/min of this volume is drawn into the deposition channel and fractionated. The design of the centrifuge minimizes inlet losses and plugging by highly agglomerated aerosols. The foil is pre-scored with a stylus so that narrow segments of foil can be separated and examined in a scanning electron microscope to determine the average equivalent volume diameter. The latter is facilitated by a set of standard SEM photographs of spheres where each sphere differs in volume from its neighbors by a factor of two. These data are used to calculate α as a function of particle size. To determine ρ_p and χ , SEM photographs are used to calculate the total number of particles on each segment of foil and then aerosol mass

is determined on each segment by flame photometry for sodium. A gravimetrically determined calibration between sodium mass and aerosol mass was obtained by weighing filter samples and then analyzing the deposit for total sodium. The sodium mass on the foil segments may be estimated by interpolation from measurements of adjoining segments as well as by direct measurement of sodium on the SEM foil section following SEM examination.

CALIBRATION PROCEDURE

The aerodynamic size calibration of the centrifuge was determined using monodisperse polystyrene latex and polyvinyl toluene spheres of known size and density. Suspensions of these spheres were aerosolized using a compressed air nebulizer and dried by mixing with dry air. The suspension concentration was adjusted to produce high concentrations of singlet, doublet, and triplet particles whose aerodynamic diameters have been determined previously.⁶

The instrument is calibrated by sampling these particles for one to two hours. This produces visible bands on the deposition foil corresponding to singlet, doublet, etc. particles. The position of these bands is measured relative to the deposition channel inlet to produce the calibration curve shown in figure 2.

An absolute calibration of the SEM magnification and the associated photographic enlargement process has been made with replicas of diffraction gratings having 54,864 lines per inch and making direct measurements of the line spacing on the final photographic enlargements.

RESULTS

Measurements have been made for one pound sodium burns in our 90 m³ chamber at ambient temperature and relative humidity ranging from 14-22%. The airborne mass concentration versus time profiles for these runs are shown in figure 3. Photographs of the fractionated aerosol for run R-11 at an aerodynamic diameter of 3.4, 1.7 and 0.8 μ m are shown in figures 4, 5, and 6. These figures show the change in agglomerated structure of the sodium aerosol with changing aerodynamic diameter.

Preliminary measurements have been made on three centrifuge samples that were taken after burning one pound of sodium in our 90 m³ chamber. The data show high variability because of the small number of particles counted and sized as well as other uncertainties. Consequently, the results shown in Table 1 are an average for the three runs. The average density modification factor, α , particle density, ρ_p , and dynamic shape factor, χ , are given for three aerodynamic diameters, 3.4, 1.7, and 0.8 μ m.

The density modification factor, α , is based on an assumed

material density of 2.74 gm/m^3 , the value used in reference 7 for the design basis accident for evaluation of emergency air cleaning systems for single containment systems. These very preliminary values for α fall between the assumed values used by HEDL⁷ and NRC⁴ of 0.25 and 0.1, respectively.

As expected, the value for particle density is smallest for the highly agglomerated large sized particles and largest for the $0.8 \text{ }\mu\text{m}$ aerodynamic particles which appear to be unitary particles. These values for particle density are believed to be high because of inaccuracies in counting and mass measurements. As a consequence, the dynamic shape factor values are also high. However, the trend is in the expected direction. The greatest χ value is for the $1.7 \text{ }\mu\text{m}$ aerodynamic diameter particles because, as shown in figure 5, these partially agglomerated particles show the greatest departure from spherical shape.

Steps have been taken to eliminate the problems that emerged during these runs. During the next period, many photos will be taken at lower magnification so that much greater numbers of particles can be counted. A more refined way of segmenting the foil will provide more reliable mass information. The equivalent volume diameter determinations will be improved by using a new set of standard sphere photographs and by averaging values according to volume rather than arithmetically.

Table 1

SUMMARY OF RESULTS

Average Values for 3 Samples Taken at the End of
One Pound Sodium Burns

	Aerodynamic Diameter		
	3.4 μm	1.7 μm	0.8 μm
Density modification factor (α) ($\rho_M = 2.74 \text{ gm/cm}^3$)	0.14	0.22	0.17
Particle density (ρ_p) gm/cm^3	0.45	1.6	2.6
Dynamic shape factor (χ)	1.4	2.5	1.8

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FIGURE 1
 DIAGRAM OF AEROSOL CENTRIFUGE

FIGURE 2

AEROSOL CENTRIFUGE CALIBRATION

FIGURE 5

BASELINE CONCENTRATION DECAY PROFILES FOR 1 LB. SODIUM

← 10 μm →

FIGURE 4

SEM PHOTOGRAPHS OF SODIUM FUME PARTICLES, AERODYNAMIC DIAMETER = 3.4 μm

10 μm

FIGURE 5

SEM PHOTOGRAPHS OF SODIUM FUME PARTICLES, AERODYNAMIC DIAMETER = 1.7 μm

← 10 μm →

FIGURE 6

SEM PHOTOGRAPHS OF SODIUM FUME PARTICLES, AERODYNAMIC DIAMETER = $0.80\ \mu\text{m}$