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(54) **Measurement of particle size distribution and mass concentration of nuclear fuel aerosols**

(57) The particle size distribution and particle mass concentration of a nuclear fuel aerosol is measured by admitting the aerosol into a vertically-extending container (1), positioning an alpha particle detector (4) within the container so that its window (4b) is horizontal and directed vertically, stopping the admission of aerosol into the container, detecting the alpha-activity of the particles of the aerosol sedimenting onto the detector window (for example in a series of equal time intervals until a constant level is reached), and converting the alpha-activity measurements into particle size distribution and/or particle mass concentration measurements. The detector 4 is attached to a pivotted arm (5) and by raising a counterweight 7 can be lowered from the container (1) for cleaning.

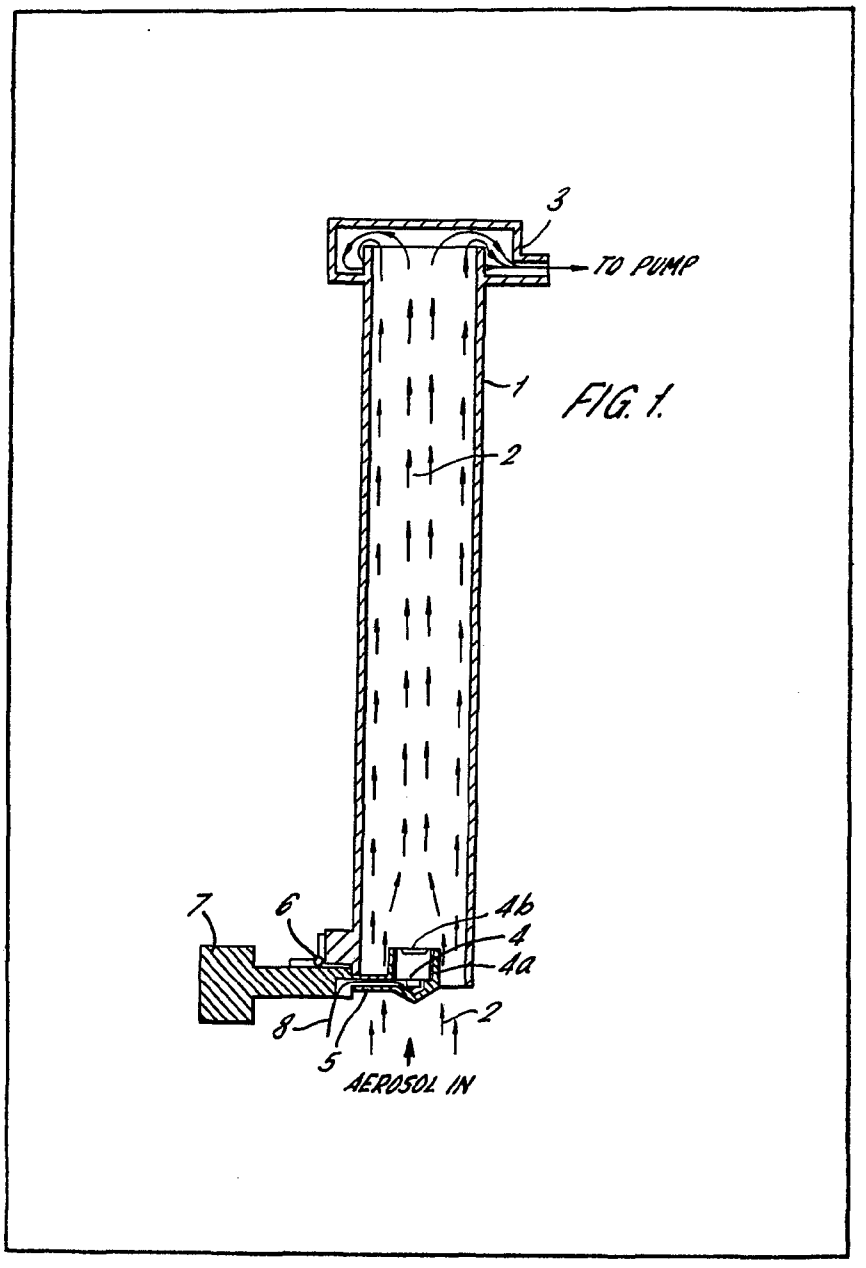

FIG. 1.

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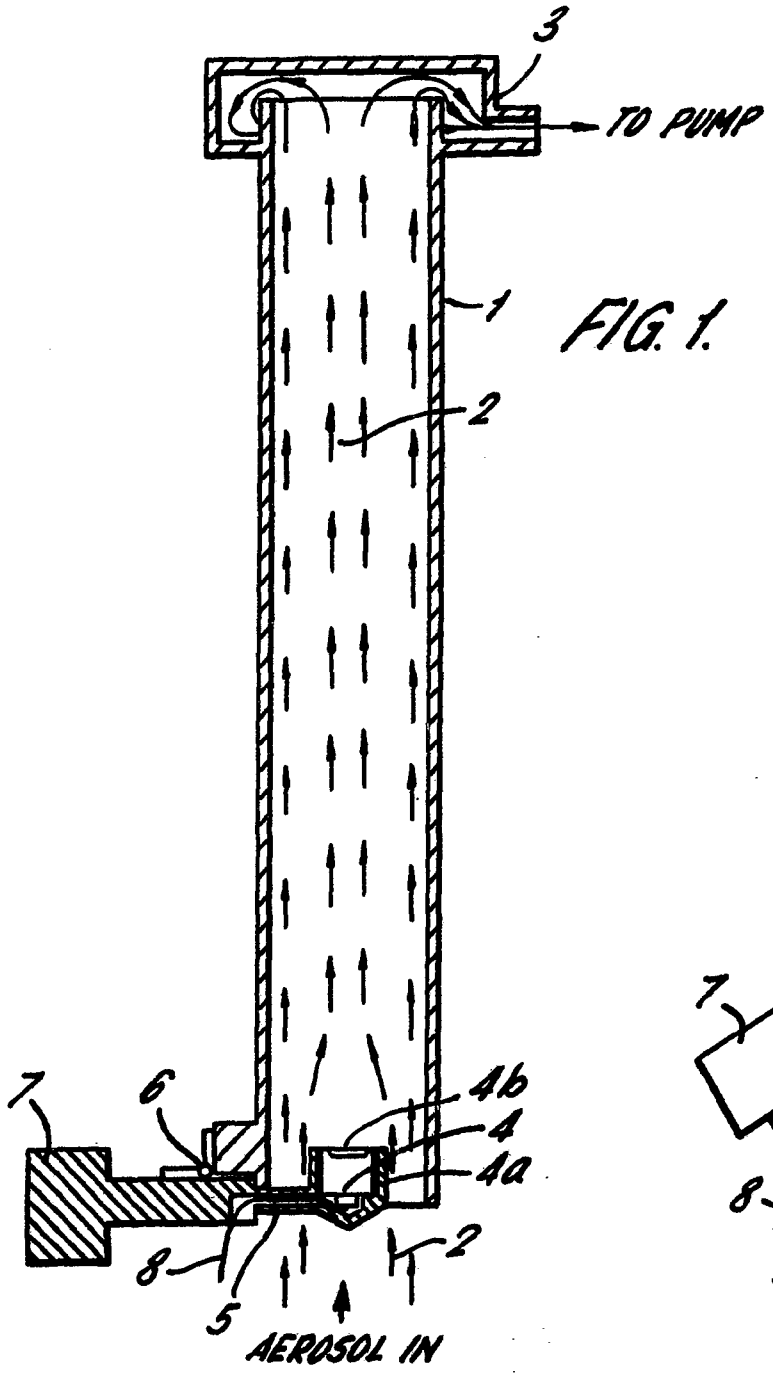


FIG. 1.

FIG. 1a.

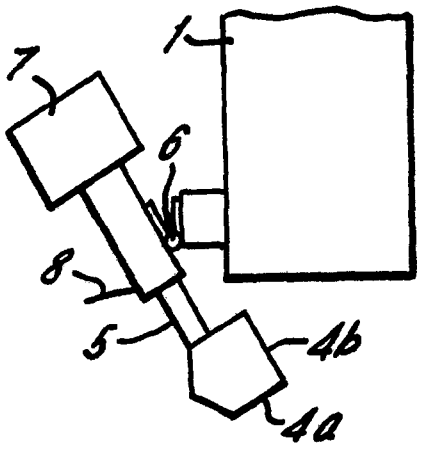


FIG. 2.

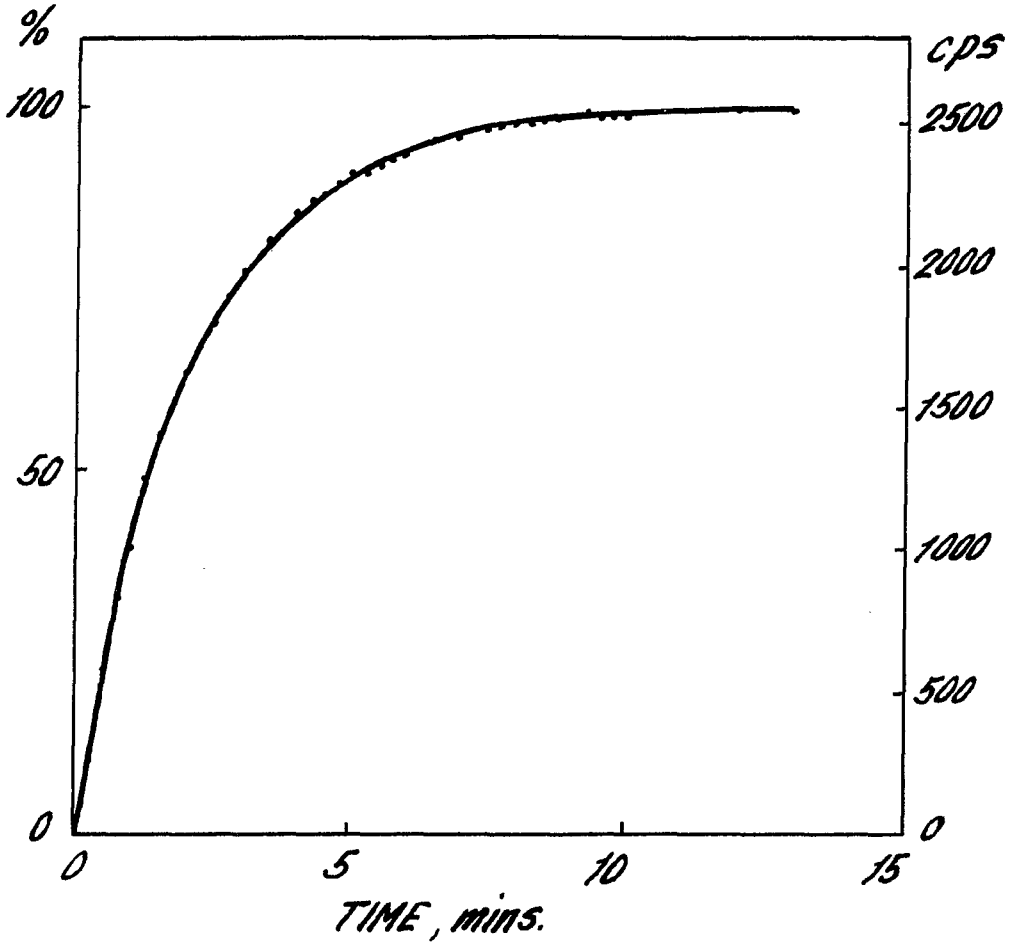


FIG. 3A.

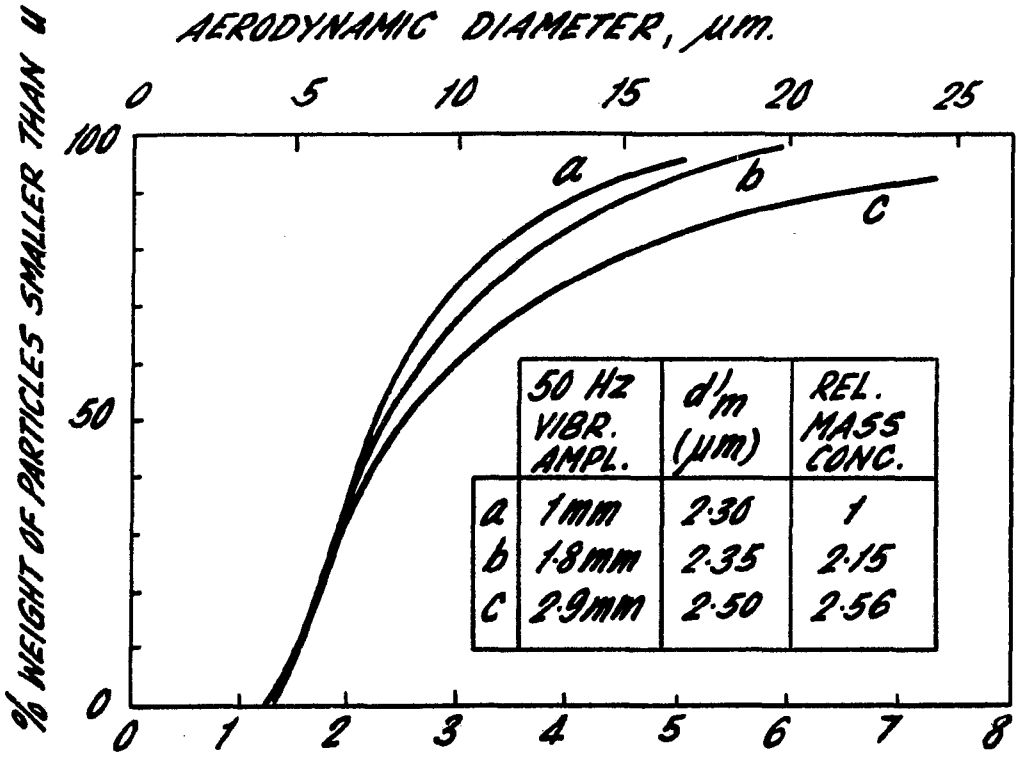

AERODYNAMIC DIAMETER, μm.
0 5 10 15 20 25
% WEIGHT OF PARTICLES SMALLER THAN W
100
50
0
0 1 2 3 4 5 6 7 8
a
b
c
50 Hz VIBR. AMPL.
d'm (μm)
REL. MASS CONC.
a 1mm 2·30 1
b 1·8mm 2·35 2·15
c 2·9mm 2·50 2·56

FIG. 3B.

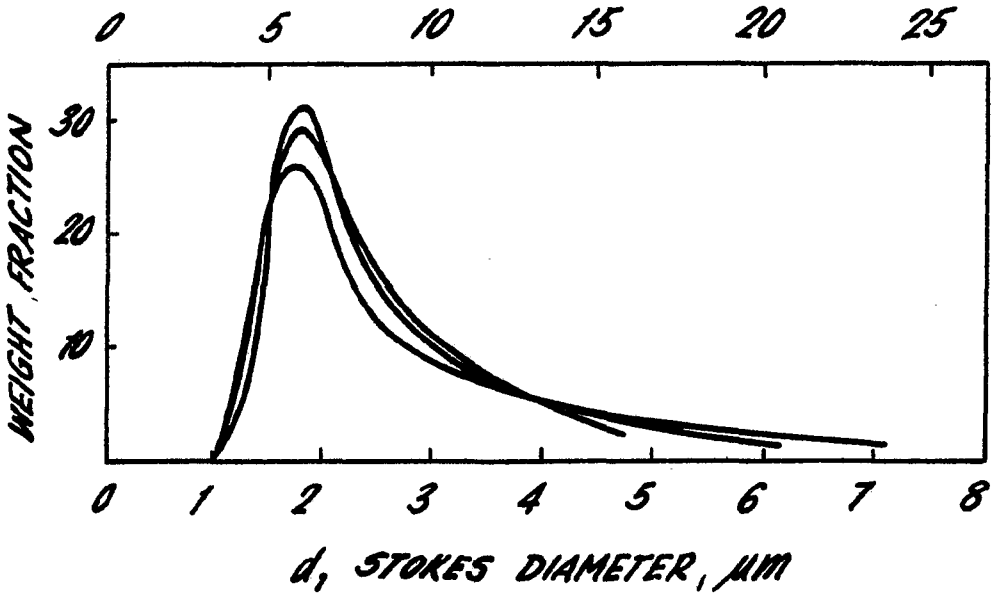
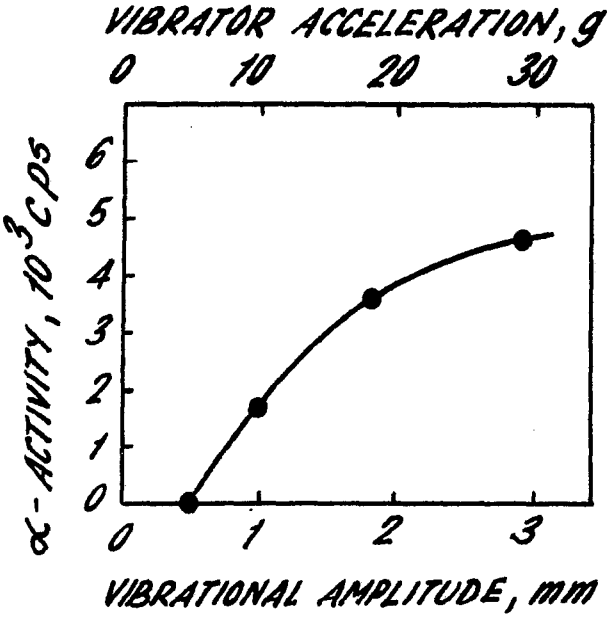

0 5 10 15 20 25
WEIGHT FRACTION
30
20
10
0 1 2 3 4 5 6 7 8
d, STOKES DIAMETER, μm

FIG. 4.


VIBRATOR ACCELERATION, g
0
10
20
30
α-ACTIVITY, 10^3 cps
0
1
2
3
4
5
6
0
1
2
3
VIBRATIONAL AMPLITUDE, mm

SPECIFICATION

Particle size distribution and particle mass concentration measurement apparatus and method for nuclear fuel aerosols

The present invention relates to an apparatus for measuring the particle size distribution and particle mass concentrations of nuclear fuel aerosols, and to a measurement method using such apparatus.

Various types of measurement apparatus are known, but each has certain disadvantages:—

a) The aerosol centrifuge

Here the aerosol enters the centre of a spiral duct cut into a rotor turning at speeds up to 3000 rpm. Large particles deposit on the duct wall near the aerosol inlet; whilst smaller particles deposit further down the duct where the centripetal acceleration is greater. In this way particles are separated into a spectrum according to their aerodynamic and inertial properties. (See W. Stöber, H. Flachsbart "Size separating precipitation of aerosols in a spinning spiral duct" Env. Sci. Tech *3* (1966)1280—1296). With this apparatus, however, the mass concentration is not measured, particles larger than 5μm cannot be separated, particle losses in the centrifuge inlet are large and depend on particle size, and the user must devise his own means of determining the particle size distribution from the deposit on the duct wall.

b) The inertial spectrometer

Here an aerosol stream, surrounded by a carrier gas, is led through a sharp 90° bend and is then drawn through a filter where aerosol particles are trapped. While passing through the 90° bend, large particles slip relative to the gas flow more than small particles, so that there is a separation of particles on the filter according to their inertial properties. (See V. Prodi et al. "An inertial spectrometer for aerosol particles" J. Aerosol Sci. *101* (1979) 411—419).

With this apparatus, however, the user must devise his own means of determining the particle size distribution from the deposit on the filter, the mass concentration is not measured, and the very fine aerosol inlet is susceptible to partial blocking, which results in a distorted aerosol deposition pattern.

c) The horizontal elutriator

Here an aerosol stream is drawn through a narrow lateral slit in the roof of a horizontal duct about 5mm high, about 50mm wide and about 600mm long. As the aerosol is carried along the duct by an air flow, aerosol particles settle out onto the floor of the duct as speeds proportional to their aerodynamic diameters; thus achieving a size separation of the aerosol particles. (See W. Stöber "Zur Bestimmung von Teilchengrößenverteilungen mit einem Horizontal-Elutriator" Staub *24* (1964) 221—223).

With this apparatus, however, the aerosol sampling rate is very low, the whole apparatus must be thermally insulated to minimize convection currents in the duct, the user must devise his own method of obtaining the particle size distribution from the deposit on the duct floor, and the mass concentration is not measured.

d) The optical counter

Here an intense beam of light is focussed onto a stream of aerosol particles. Aerosol particles passing through the focussed beam create pulses of scattered light which are measured by a photodetector. The size of each scattered light pulse and the signal amplitude of the detector are proportional to the aerosol particle size. (See K. T. Whitby, K. Willeke "Single Particle Optical Counters: Principles and Field Use" p. 145—182 in "Aerosol Measurement" (1979), University of Florida Press.)

With this apparatus, however, the amount of light scattered from irregularly-shaped particles depends on the orientation of the particles, partly-illuminated particles increase the apparent concentration of small particles; whilst coincident illumination of particles increases the apparent concentration of large particles, and the apparatus is expensive.

e) The sedimentation microbalance

Here a sample of powder is first suspended in a liquid and is then allowed to settle out onto the immersed pan of a microbalance. A particle size distribution can be derived from the resulting cumulative weight-time curve with the aid of Stokes Law. (See S. Oden, Proc. Roy. Soc. Edinb. 36 (1915) 219, A. E. Jacobsen, W. F. Sullivan, Ind, Eng. Chem. *19* (1947) 855 and W. Bostock, J. Sci. Instr. *29* (1952) 209).

With this apparatus, however, particles below about 5μm cannot be measured easily, and a high mass concentration of particles is required, so that this apparatus is suited more to characterizing bulk powders than to aerosol measurement.

In accordance with the present invention there is provided an apparatus for measuring the particle size distribution and particle mass concentration of a nuclear fuel aerosol comprising a vertically-extending container for the aerosol, means for admitting the aerosol into the container, and an alpha-particle detector positioned or positionable within the container and having a detector window disposed horizontally and directed vertically, so as to detect the alpha-activity of particles of the aerosol sedimenting onto the window.

The present invention also provides a method of measuring the particle size distribution and particle mass concentration of a nuclear fuel aerosol, which method comprises admitting the aerosol into a vertically-extending container for the aerosol, positioning an alpha particle detector within the container so that its detector window is horizontal and directed vertically, stopping the

admission of aerosols into the container, detecting the alpha-activity of the particles of the aerosol sedimenting onto the detector window, and converting the alpha-activity measurement into particle size distribution and/or particle mass concentration measurements.

In the apparatus of the present invention a cloud of nuclear fuel aerosol is allowed to settle onto an alpha particle detector. As the aerosol particles settle onto the detector the measured alpha-activity increases. Analysis of the alpha-activity-time curve in a manner similar to that used for the sedimentation microbalance enables the particle size distribution to be calculated. The use of an alpha-detector greatly increases the sensitivity of the method compared with previous sedimentation microbalances, so that direct measurements are possible on nuclear fuel aerosol clouds containing particles as small as 1 micron.

One embodiment of the present invention will now be described by way of example, with reference to the accompanying drawings, in which:—

Figure 1 is a cross-sectional view of the apparatus of the present invention taken along a plane perpendicular to the hinge axis and along the axis of the tube; and apparatus being in its operative position;

Figure 1a is a schematic view of the apparatus of Figure 1 in its inoperative, cleaning position;

Figure 2 is a graph showing the variation of alpha activity with time as a result of aerosol sedimentation;

Figures 3A and 3B are graphs showing particle size distributions of $UPuO_2$ aerosols on a cumulative weight and a single weight basis, respectively; and

Figure 4 is a graph showing the variation in yield of $UPuO_2$ aerosol from fuel pellets.

In Figure 1, the apparatus consists of a circular tube, 1, approximately 400mm high and 60mm in diameter through which the aerosol, 2, is drawn by a pump (not shown) connected to outlet 3. A commercially-available alpha-detector, 4, is mounted in the centre of the aerosol stream on an arm, 5, connected to tube, 1, by a hinge, 6. The detector 4, is held in an alpha-radiation-resistant housing, 4a, and is of the "surface barrier" type. The housing 4a, has a surface barrier, 4b, forming a detector window, which barrier is arranged to lie horizontally and be directed vertically, when the detector is in its operative position, so that particles of the aerosol can sediment onto the window and their alpha-activity can be detected. The arm, 5, has a counterweight, 7 which ensures that the alpha-detector, 4, is normally held face-upward in tube 1. By raising the counterweight, 7, as shown in Figure 1a, the detector, 4, can be conveniently lowered for cleaning with a jet (not shown) of compressed gas. The alpha-detector 4, is connected by a coaxial cable, 8, to conventional alpha-counting equipment (not shown).

The measuring method using the present apparatus is as follows:—

The sampling pump (not shown) is switched on and the aerosol is drawn up through the tube, 1, for about 2 mins. at a flow rate of about 10 l/min. The pump is then switched off and the aerosol in the tube begins to settle under gravity. Alpha counting is started shortly before the pump is switching off and the alpha-activity is recorded over successive short time intervals (typically 1 to 10s) until a constant alpha-activity level is recorded (typically 15 to 20 mins). Recording the alpha activity in this way effectively gives the differential of the cumulative activity-time curve. The differential of the cumulative activity-time curve is equivalent to a cumulative weight-time plot and the data may be evaluated with techniques developed for the sedimentation balance e.g. Odens method of tangential intercepts. The final activity measured on the detector is proportional to the mass of particles on the detector, so that the mass concentration of the particles in the aerosol may also be calculated if the isotopic composition of the aerosol and the detector efficiency are known.

Some advantages of the described apparatus and method are that results can be obtained rapidly, particularly if an interfaced desk calculator is used to handle the data, the weight distribution of the aerosol particles can be obtained directly as a function of both the Stokes diameter and the aerodynamic diameter of the particles, the mass concentration may also be measured for materials of known isotopic composition, and the apparatus is simple, relatively inexpensive, robust and easy to operate under the remote handling conditions necessary for nuclear fuels, e.g. in their manufacture, use and reprocessing.

Example

A trial was carried out using the described apparatus in which the nuclear fuel aerosol was generated by vibrating a perforated aluminium pot containing UP_uO_2 fuel pellets in the gas stream upstream of the apparatus. The aerosol generator was started and the gas was drawn upwards by the pump at 19mm/s to carry a stream of aerosol particles past the detector. Once a steady flow was established (3 mins) the generator and gas flow were stopped and alpha-counting was started. As the aerosol particles in the tube above the detector settles out, the alpha-activity recorded by the detector increased with time as shown in Figure 2.

Provided that no significant self absorption of alpha-particles occurred within the aerosol sediment on the detector, the recorded alpha-activity could be assumed to be directly proportional to the mass of the sediment. The activity-time curve of Figure 2 was thus equivalent to a cumulative weight-time curve and the known mathematical treatments developed for sedimentation balances could be applied.

The measured activity-time curves were treated using Oden's method of tangential intercepts, i.e.:

$100\int_0^D F(D)dD = 100$—tangential intercept (1)

where F(D)dD=the weight fraction of particles with a diameter between D and D+dD and where the tangential intercept is the intercept of the tangent of the activity-time curve with the y axis expressed as % of the final activity.

The correspondence of settling time to particle diameter was obtained by equating the frictional force (Stokes Law) to the gravitational force on the particles, i.e.:

$$D = \sqrt{\frac{18\,\eta\,h}{\rho\,g\,t}} \qquad (2)$$

D=the Stokes diameter of the particle
n=the viscosity of the aerosol gas
g=the gravitational acceleration constant
e=the density of the aerosol particle material
t=the settling time
h=the settling distance

Plotting $\int_0^D F(D)dD$ against D gave the cumulative weight distribution as shown in Figure 3A, from which a simple weight distribution curve could be obtained if required as shown in Figure 3B. The weight distribution as a function of aerodynamic diameter was obtained simply by multiplying the Stokes diameter, D, by $\sqrt{e}$. The tangent intercepts of the activity-time curves could be obtained most accurately when the curvature of the plots was greatest. Maximum curvature corresponds to the range of particle diameters where the mass fraction is highest i.e. to the peak of the simple weight distribution curve and thus to the most interesting portion of the size distribution. In contrast, the mass fraction at the upper and lower ends of the size distribution is low and thus the accuracy of the weight distribution is lower than at the peak.

Because the recorded alpha-activity is proportional to the mass of the settled aerosol particles, the mass concentration of particles in the aerosol could be calculated, provided that the isotopic composition of the aerosol material and the detector efficiency were known.

Sedimentation experiments were made with aerosols generated by vibrating sintered $UPuO_2$ pellets at various amplitudes. The typical alpha-activity-time curve shown in Figure 2 is the average of 4 successive experiments under identical conditions. The reproducibility of the data was found to be satisfactory. Using Odens method of tangential intercepts, it was found that the mass median particle diameter increased slightly with increasing vibrational amplitude from 2.3 to 2.5 μm over the range investigated. The lowest vibrational amplitude (curve a in Figures 3A and 3B) produced aerosols with an approximately log-normal particle size distribution (σ=1.4 μm). With increasing vibrational amplitude however, the proportion of large particles increased so that the distributions were positively skewed rather than log-normal (curves b and c in Figures 3A and 3B.

As shown in Figure 4, the quantity of aerosol produced, as measured by the final alpha-activity on the counter, was found to increase with vibrational amplitude once a threshold corresponding to a maximum acceleration of 5g was exceeded.

Throughout this specification, the word "aerosol" has been used. It is not intended however, that the invention should be limited to the use of a system of "colloidal" particles. Any particle dispersion in a carrier fluid can be used; the preferred fluid being a gas, rather than a liquid.

Claims

1. An apparatus for measuring the particle size distribution and partial mass concentration of a nuclear fuel aerosol comprising a vertically-extending container for the aerosol, means for admitting the aerosol into the container, and an alpha-particle detector positioned or positionable within the container and having a detector window disposed horizontally and directed vertically, so as to detect the alpha-activity of particles of the aerosol sedimenting onto the window.

2. An apparatus as claimed in claim 1 wherein the container is in the form of a tube whose axis is vertical, and wherein the means for admitting the aerosol into the container is a suction pump connected to the upper end of the tube.

3. An apparatus as claimed in claim 1 or claim 2 wherein means are provided for cleaning the detector window of the detector between measurements.

4. An apparatus as claimed in claim 3 wherein the detector is movable within the container from a measurement station to a cleaning station.

5. An apparatus as claimed in claim 3 or claim 4 wherein the cleaning means comprises a jet of gas.

6. An apparatus as claimed in claim 1 substantially as hereinbefore described with reference to and as illustrated in Figures 1 and 1a of the accompanying drawings.

7. A method of measuring the particle size distribution and particle mass concentration of a nuclear fuel aerosol, which method comprises admitting the aerosol into a vertically-extending container for the aerosol, positioning an alpha particle detector within the container so that its detector window is horizontal and directed vertically, stopping the admission of aerosol into the container, detecting the alpha-activity of the particles of the aerosol sedimenting onto the detector window, and converting the alpha-activity measurements into particle size distribution and/or particle mass concentration measurements.

8. A method as claimed in claim 7 wherein the detecting step is commenced before the admission of aerosol is stopped.

9. A method as claimed in claim 7 or claim 8 wherein the alpha-activity of the sedimenting particles is recorded over successive, relatively

short time intervals until a constant alpha-activity level is recorded, so as to provide the differential of the cumulative alpha-activity/time curve.

10. A method as claimed in any one of claims 7 to 9 wherein the method is repeated several times in order to provide an average measurement, with the detector window of the detector being cleaned with a jet of gas between measurements.

11. A method as claimed in claim 7 substantially as hereinbefore described.

12. A method as claimed in claim 7 substantially as hereinbefore described in the specific Example.

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