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EVALUATION OF AN EXPLODED-WIRE AEROSOL GENERATOR FOR USE IN INHALATION STUDIES

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Abstract—An exploded wire aerosol generator was evaluated using test aerosols derived from silver wires and results were compared with existing published information on exploded wire particles. Aerosol properties relevant to inhalation studies were emphasized. Primary particles were spherical, metallic, and distributed nearly lognormally by volume (geometric standard deviation about 1.5) with count median diameters between 0.05 and 0.008 μm. The primary particle size decreased as the explosion energy per unit mass of wire was increased. Primary particles rapidly formed agglomerates of up to hundreds of spherical units but an air blast aimed at and synchronized with the wire explosion was successful in reducing agglomeration. Agglomerate aerosols with submicron aerodynamic diameters were produced by rapidly diluting the explosion zone after the air blast. Agglomerates of 0.026 μm count median diameter primary particles had measured specific surfaces of 16.4 m²/g and solubility rate constants of 10 μg/cm² per day in a lung fluid simulant containing protein. The relevance of exploded wire agglomerates to inhalation studies is briefly discussed.

INTRODUCTION

The electrical explosion of a wire, or any other conductor, has two necessary conditions. First, the energy input per unit mass of material must exceed that necessary for vaporization, and second the rate of energy input must be great enough to produce an explosive expansion. The end product of a wire explosion, the debris or smoke, is the subject of this investigation.

A basic generator for producing aerosols by exploding wires is schematized in Fig. 1. A large capacitor, initially charged to several thousand volts, is discharged through the wire by closing S. Assuming typical values of capacitance, inductance, initial voltage, wire mass etc., the wire is heated to many times its vaporization temperature by the current surge before it undergoes any significant change in shape. The wire behaves as a superheated vapor, explosively expands outward and, if the voltage is great enough, ionizes forming a plasma cloud. A few microseconds after the switch is closed the conduction phase ends and cooling proceeds. An aerosol forms and diffuses throughout the chamber. Exploded wire (EW) aerosols consist of particles that are nearly always smooth, spherical and less than 0.1 μm in diameter. Descriptions of this explosion process have been given by many investigators (Bennett et al., 1962; Funfer et al., 1958; Mel'nikov and Barchenko, 1965).

The energy originally stored in the capacitor, E = 1/2 C(V)², goes into circuit losses, radiant emissions, shock waves, ionization and excitation of metal and air, wire expansion and the creation of new surface area (Yakovlev, 1969). The partition of energy varies with time and specific conditions, but excellent reproducibility is observed from explosion to explosion if the initial conditions remain fixed.

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This investigation was designed to study the properties of EW aerosols that are relevant to inhalation studies; viz particle composition, shape, size distribution, agglomeration, solubility, specific surface and aerodynamic properties.

PREVIOUS WORK

An abundant literature on exploding conductor phenomena can be obtained from the bibliographies of CHACE (1958) and CHACE and WATSON (1962, 1965, 1967). However a review of this literature reveals few direct references to exploded-conductor particles; nearly all of the quantitative information on EW aerosols is widely scattered.

In perhaps the earliest EW-aerosol study, ABRAMS (1946) compared several methods of producing radioactive aerosols, including exploding wires and foils. They worked with a 1.5 μF capacitor bank charged to 35 kV or less, and exploded wires, foils and pieces of Al, U and Pu metal. Agglomerates of spherical particles were observed by electron microscopy. The mean particle diameter was given as 0.2 μm (presumably an agglomerate size) with all chains less than 1 μm in length.

![Fig. 1. Simplified exploding wire aerosol generator. C low inductance capacitor; HV high voltage charging supply; S switch; W wire inside explosion chamber E.](image)

Using electron microscopy and electron- and X-ray diffraction techniques, HARVEY et al. (1960) examined the structure and form of electric arc smokes. Though most of their data were on particles generated by evaporating various metals from tungsten wires, they did publish information on particles from Ag and Au exploded wires. The EW particles were metallic, "globular" (some had weakly developed crystal faces), and 0.03 – 0.065 μm in diameter. Since particle size measurements by electron microscopy and X-ray crystallography agreed, they concluded that EW particles were single crystals.

Data on zirconium particles formed in wire explosions under water were presented by BAKER and WARCHAL (1962). They used a 100 μF capacitor bank charged to 5 kV or less to explode 30 and 60 mil wires. Some of their experiments involved only melting of the wires, but spherical particles were always observed when there was an explosive pressure rise. The energy per unit mass of wire was uncommonly small (0.42 – 1.65 j/mg) in this study. The mean Sauter diameter* of the zirconium particles (2280 – 100 μm) tended to decrease with increasing explosion energy.

*Mean Sauter diameter = Σnd^3/Σnd^2, where n particles have diameter d.
Coagulation measurements on EW particles were made by Rosinski et al. (1962). They found that the coagulation constant was influenced by the specific radioactivity of exploded Ag, Au and Cd wires. Mean values of the coagulation constants were $0.54$ and $0.51 \times 10^{-9}$ cm$^3$/sec for nonradioactive Ag and Cd, respectively, and between about $0.5$ and $3 \times 10^{-9}$ cm$^3$/sec for various specific activities of Au. For increasing specific activity, the coagulation constant was observed to decrease and then to increase above that for nonradioactive aerosols.

Considerable information on EW aerosols generated from 15 metals with a 20 $\mu$F capacitor bank charged to 2 – 20 kV has been published by Karioris and Fish (1962), Karioris et al. (1962), Karioris and Wyci (1963), Karioris, Fish and Moll (1963) and Barkow, Karioris and Stoffels (1963). Most of the metals they exploded in air yielded metals mixed with metal oxides. No non-metallic forms, using X-ray diffraction analysis were found for Ag, Au and Pt in air of for Ag, Au, Mo, Pt and Sn exploded in nitrogen. Yield studies indicated that above a critical voltage most of the metal was converted into recoverable aerosol. Primary particles were spherical, crystalline and had mean crystal diameters of 0.005 – 0.2 $\mu$m (determined by X-ray line broadening). In agreement with Harvey et al. (1960), particle sizes by electron microscopy were comparable to mean crystal diameters. The primary particle size distribution (by count) was described as between normal and lognormal and some bimodal size distributions were presented. A decrease in particle and crystal diameter was generally seen as the explosion voltage, and hence the energy per unit mass of wire, was increased. Agglomeration occurred, but they reported that individual primary spheres could be obtained by fanning the aerosol until all agglomerates were collected on the walls of a plenum chamber. Using a nitrogen adsorption technique, the specific surface of a sample of EW U$_3$O$_8$ particles was found to be 15.2 m$^2$/g.

Couchman (1965) generated particles from Al, Au, Ni and Pt wires with a current surge from an 11 $\mu$F, 25 kV capacitor bank. His object was to study the effect of particle density on cascade impactor separation. Though his technique did not always lead to a wire explosion, he described particles as small as 0.01 $\mu$m in diameter from some exploded wires. The particles were extensively agglomerated spheres that had the same density as the unexploded wires. It is worth noting that several of his aerosols were contaminated with what seemed to be explosion debris from the titanium wire supports.

The charging and mobility of EW Cu agglomerates of 10 – 300 primary spheres was examined by Vomela (1966) and Vomela and Whitby (1967). They reported that a capacitor discharge of 810 J produced primary particles having a mass median diameter (NMD) of 0.043 $\mu$m and a geometric standard deviation of about 1.3, implying that the mass distribution was lognormal with a count (number) median diameter (CMD) of about 0.035 $\mu$m.

Particle size measurements during the explosion of Cu wires (31.7 $\mu$F capacitor at 3 kV) were attempted by Weber and Shear (1969). They looked at the ratio of monochromatic light (4358 Å) scattered at 45° and 135° vs time during the explosion. They reported that the particle size reached a maximum early in the explosion and then dropped to an immeasurably small value (less than 0.01 $\mu$m) by the time of the explosion voltage peak. There were some serious experimental problems, but a mean particle size of 0.12 ± 0.035 $\mu$m was determined. Although presented as such, it is not clear that this size measurement applies to the solid particles collected after a wire explosion.

For those cases cited, where enough information was published to do so, a plot of particle CMD versus explosion energy per unit mass of wire is shown (Fig. 2.). The particle
size results of this study, along with an unpublished result obtained by the author in 1966, are also shown on the same figure. Where the CMD was not possible to estimate, the mean crystal size was plotted. This parameter should probably be interpreted as a mean particle volume (Peiser et al., 1960). Since no information on the dispersion of particle sizes is obtained from X-ray line broadening measurements, the CMD could not be calculated from the crystal size data. The figure shows that primary particle sizes tend to fall within relatively narrow limits, and decrease with increasing explosion energy per unit mass of wire.

![Image of graph showing particle diameter vs. explosion energy per unit mass of wire](https://example.com/graph.png)

**Fig. 2.** Dependence of particle diameter on explosion energy per unit mass of wire. △ Phalen, CMD, Ag in air; [] Vomela and Whittby (1967), CMD, Cu in air; ⫻ Barkow et al. (1963), crystal size, Cu in air; ○ Karioris and Woyci (1963), crystal size, Au, Cu and Mo in nitrogen; ◇ Weber and Shear (1969), light scatter, Cu in air.

**EXPERIMENTAL**

The EW aerosol generator was constructed in three sections: the electronic circuitry, the containment or explosion chamber, and the dilution chamber (Fig. 3). The circuit consisted primarily of a 6600 J, low inductance (less than 1 μH) energy storage capacitor (Sprague type 409P, 23 μF, 24 kV), a 0 – 30 kV charging supply, a triggered sphere gap switch, a pair of tungsten wire support electrodes and two resistor banks, one to prevent inadvertent charging of the capacitor and the other to allow quick discharge when necessary. These resistor banks are important safety devices as the charged capacitor is a lethal shock hazard. Leads between the capacitor and the silver wire were short and heavy to minimize inductance and resistance.
The explosion chamber, a grounded 55 gal steel drum, was fitted with electrical leads from the capacitor, ports for aerosol sampling, an outlet to the dilution chamber, a filter barrier to the lab air and a disposable aluminium foil liner.

A grounded 40 l. spirometer partially filled with clean air was used to withdraw and dilute a portion of the aerosol from the explosion chamber. This served to slow agglomeration and to store a relatively stabilized aerosol for further study or use. The criterion for stability was that the median aerodynamic diameter of the aerosol change less than 10 per cent in a 15 min period. This criterion was chosen as it is convenient for brief inhalation studies.
Silver wires of better than 99.99 per cent purity were exploded throughout this study. Wires were 0.5 - 12.5 cm long and 5, 7 or 9 mils in diameter so that the mass exploded ranged from fractions of a milligram to about 50 mg. Neutron activated wires (Ag 109 + n → Ag 110m, 255 day half life, β, γ) with a specific activity of about 30 μCi/mg were used for the aerodynamic and solubility measurements. The silver wire was sealed in nitrogen filled quartz tubes before irradiation to prevent chemical decomposition under reactor conditions.

Preliminary wire explosions in open air formed a dense smoke that remained near the wire seconds for a few seconds before dispersing. Since this condition of high particle count (10⁸ to 10¹⁰/cm³) favors rapid initial agglomeration rates, an air blast was installed in the explosion chamber to disperse the initial smoke cloud. This clean air blast, of about 2 l., was directed at the wire and triggered by a solenoid valve that opened for 1 sec when the sphere gap was fired.

Aerosol samples for electron microscopy were collected on polyvinyl chloride or carbon substrates, using thermal and electrostatic precipitators. Grids were examined and photographs taken for particle sizing on a Philips EM 100B and a Philips EM 200 with a liquid nitrogen cold finger attachment. Size distributions were determined from photographic enlargements (about 200,000 X) using a Zeiss TGZ3 particle size analyser.

Agglomeration was examined by particle counting with a General Electric G1 condensation nuclei counter. Wall and sedimentation losses were ignored during the first several minutes and the decrease in particle number was attributed solely to agglomeration. A strip chart recording of the time variation of number of nuclei/cm³ was used to estimate the coagulation constant, c, (cm³/sec) and to evaluate the effectiveness of the air blast in slowing initial agglomeration.

Aerodynamic size distributions of agglomerates were obtained from the explosion and dilution chambers with low-flow-rate, seven-stage, cascade impactors (Mercer et al., 1970). The impactors were operated at flow rates so that at least three stages had cutoff diameters between 0.1 and 1 μm the region of major interest in this study.

Surface areas were obtained from BET plots of nitrogen adsorption data obtained using a Sartorius 4102 electric microbalance.

Samples for X-ray analysis collected from the explosion chamber on membrane filters were scraped into either diffractometer sample holders or thin-walled quartz capillary tubes. X-ray analysis was done on a General Electric diffractometer and with a 7.16 cm radius Debye-Scherrer camera. Nickel filtered Cu Kα radiation (1.54 Å) was used for all measurements.

Solubility measurements were made by radioactivity counting of filtered aliquots taken at various times from a stirred mixture of aerosol and solvent (Morrow et al., 1971). Particles were placed in 420 - 500 cm³ of solvent maintained at 35° - 37° C. The media were either water, an interstitial fluid simulant (after Gamble, 1952) with protein, or this fluid simulant with acetate ion in equimolar substitution for the protein. The determination of a solubility rate constant, k, in gm/cm² per day, was the object of these measurements.

### Results and Discussion

Fresh samples of exploded silver wires collected on membrane filters were black and velvety in appearance. X-ray diffraction examination of this material indicated that metallic...
Fig. 4. Debye-Scherrer patterns of pure Ag wire (above) and the aerosol from exploded Ag wire (below).

Fig. 5. Electron microscope photographs of the aerosol from Ag wires exploded in air. (A) Chain agglomerates, Cr shadowed at about 30°. (B) Agglomerate as seen in electron microscope equipped with vapor suppression device. (C) Same agglomerate as in B about 1 min after removal of vapor suppression device.
silver was the only detectable crystalline material present (Fig. 4). It is not possible to rule out the presence of traces of other chemical species either in amorphous states or as very small crystals since the presence of large numbers of atoms in an ordered array is required to produce identifiable scattering at Bragg angles.

The electron microscope revealed complexly-branched, chain agglomerates containing up to hundreds of spherical particles (Fig. 5). These agglomerates often showed a preferred orientation perpendicular to the substrate plane. Each primary particle appeared to be a smooth sphere of uniform opacity. At higher magnification an internal banding, presumably due to scattering of the electron beam inside the particles, was observed (Fig. 5B).

Unless the particles were viewed in an electron microscope possessing a column vapor-suppression devise, such as a liquid nitrogen cold finger, they acquired a contaminant coating within seconds. The agglomerate of Fig. 5B is shown in Fig. 5C 1 min after removal of a liquid nitrogen cold trap. The contaminant coating on these small particles can be thicker than the particle radius, and serious overestimates of particle sizes could result from sizing contaminated particles of low electron opacity from high contrast photographs.

The CMD of primary EW particles was found to decrease with increasing explosion energy per unit mass of wire (Fig. 6). Assuming a lognormal count distribution (which is not strictly valid for these particles) the geometric standard deviation does not seem to vary appreciably with explosion energy. The practical specific energy range for this wire exploder was between about 10 and 1000 J/mg for silver wires. Higher energies required short wires, leading to arcing between the wire supports, while lower energies failed to explode the wire. For this wire exploder the dependence of CMD on specific energy input \( I \), in J/mg, (Fig. 6) can be roughly described by the relation: \( \text{CMD (\( \mu \text{m} \))} = 0.04 \exp(-1.58 I) \). The particle size relationship for a given wire exploder would be expected to depend on the exact nature of the circuitry, and on the type of wire exploded.

The cumulative count, area and volume size distributions for 1000 primary particles (Fig. 7) show that the volume is adequately fitted by the cumulative lognormal function. The count appears to be nearly normally distributed, except of course at the extremes.
Figure 7. Size distributions of primary particles of Ag EW aerosol having a number median diameter of 0.026 μm.

(a) Cumulative distributions on log-probability paper.

Perhaps this explains why some investigators have inferred that EW particles are lognormally distributed while others have not found this to be the case. A more detailed analysis of the 1000 primary particles shows that they may be described by the sum of two lognormal functions, each representing 500 particles (Fig. 7b). This form of analysis was convenient for computational purposes but it is not intended to imply that two distinct particle generating phenomena occur during a wire explosion. The size distributions presented here are somewhat distorted as the effect of smaller particles being obscured by larger ones was not corrected for. The obscuring effect might be especially important as the agglomerates often had preferred orientation perpendicular to the grid.

(b) The number distribution as the sum of two lognormal functions.
The results of the solubilization studies are shown in Fig. 8. Ideally the initial slopes of these curves estimate the solubility rate constant $k$, in infinite or constantly renewed media. However, in a real system where the presence of an anomalous phase on the same particle surfaces delayed wetting of the sample, chemical reactions between the solvent and the sample or other similar factors might influence dissolution, it is not a trivial matter to determine $k$. The slope of a straight line fit to the data of the first 15 min was used as the upper limit for $k$, and the slope of the data between 15 and 120 min was used as the lower limit. The ranges for $k$ in $\mu g/cm^2$ per day are 0.5 - 4 for water, 3 - 10 for Gamble’s with acetate, and 10 - 25 for Gamble’s with protein. It was apparent that the presence of protein greatly facilitated the dissolution of the silver samples. That this effect was solely due to the binding of ionic silver by protein, which would decrease the rate of recondensation onto the solid, is a tenable explanation. However these data also support the hypothesis that the sample dissolved at two rates; i.e. that a small fraction of the sample was more soluble than the rest. This latter interpretation implies that the more soluble fraction had a rate constant of about 10 $\mu g/cm^2$ per day even when protein was not present and that the less soluble fraction dissolved at a rate that was significantly increased by the availability of protein. In either case the value of 10$\mu g/cm^2$ per day is likely to be the most useful in predicting dissolution in the respiratory system where the availability of protein is expected. Inhalation experiments with tracheally intubated dogs using this EW aerosol were performed. Collections of blood, liver and bile, 6 hours after an acute single exposure, accounted for nearly 5 per cent of the total deposited radioactivity. This lends support to the idea that these silver particles have an appreciable dissolution rate in the lung.

The calculated specific surface of the 1000 particles analyzed in Fig. 7 was 15.4 m$^2$/g. This value was obtained from the ratio of the summation of surface area and of mass for each interval of the Zeiss TGZ3 particle size distribution area. The difference between this value and the measured value of 16.4 m$^2$/g is within the experimental uncertainty of the
BET method. This result was consistent with the electron microscope findings of smooth particles, and is in good agreement with the value of 15.2 m²/g published for EW U₃O₈ (KARIORIS et al., 1962) generated under similar conditions.

The average coagulation constant for the freshly-generated, silver aerosol was 2 × 10⁻⁹ cm³/sec. This value is somewhat higher than those reported by ROSINSKI et al., (1962) for silver and may be due to the fact that his particles were electrically discharged prior to measurement. Assuming this coagulation constant and an initial particle concentration of 10⁹/cm³ in the vicinity of the electrodes, the half time for coagulation was about 0.5 sec. The air blast was designed to disperse the initial cloud within a few tenths of a second and thereby slow this initial agglomeration. The air blast increased the particle count, as seen in Fig. 9, indicating it was successful in delaying agglomeration. It also appears that the air blast reduced particle count fluctuations at the sampling part. This was presumably the result of more thorough mixing of the aerosol due to the air blast turbulence.

![Graph showing particle count in the explosion chamber with and without air blast.](image)

**FIG. 9.** Particle count in the explosion chamber with and without air blast.

Cascade impactor measurements indicated that a few seconds after generation (using the air blast) the aerosol had an activity median aerodynamic diameter (AMAD) of about 0.2 µm and a geometric standard deviation of 1.5. Coagulation increased the AMAD to about 0.6 µm before aerodynamic size stability was reached inside the explosion chamber.

By rapid dilution of the fresh aerosol, i.e. 1 part aerosol to 9 or more parts clean air, into the dilution chamber, stable aerosols with AMAD's as low as 0.3 µm were produced. No further efforts in reducing agglomeration were attempted as these aerosols were deemed adequate for inhalation studies.
CONCLUSIONS

The chief value of the exploding wire aerosol generator lies in its ability to produce precise amounts of submicron spheres almost instantaneously. These spherical particles are crystalline, have a nearly lognormal mass distribution, and have CMD's in the hundredth micron range, the size depending on the explosion energy per unit mass of wire.

The initial rapid agglomeration of these particles, due to their high concentration, can be slowed by an air blast and dilution system permitting the generation of aerosol clouds with stable AMAD's as small as about 0.3 μm. Such agglomerate aerosols possess several characteristics of interest to the inhalation investigator. For example, they have large specific surfaces and hence would be expected to be more chemically active in the respiratory system than their simple-shaped aerodynamic counterparts. A particle agglomerate could adsorb more gaseous material, possible toxic material, on its surface than a smooth sphere having the same mass. Additionally, such an agglomerate would have a larger viscous/gravitational force ratio, causing it to remain airborne for a longer time under usual environmental conditions. The agglomerate could be expected to dissolve faster in the respiratory system which, for many substances, would result in more material being taken into the blood for distribution to internal organs. In short, for a given material, agglomerate particles probably represent a greater overall hazard than simpler-shaped particles. As with monodisperse aerosols, the primary particle size distribution of EW agglomerate material deposited by inhalation should be identical to the primary particle size distribution found in air samples.

The high rate of dissolution for these particles in an interstitial fluid simulant is also relevant to inhalation problems. A model for estimating the dissolution of lung burdens (MERCER, 1967) was applied to the hypothetical inhalation of EW silver particles (density = 10.5 gm/cm³) having the size distribution in Fig. 7 and a solubility rate constant of 10 μg/cm² per day. Calculation indicated that half of the inhaled material would dissolve in the first 10 hr and that only about 1 per cent would remain undissolved after 2 days. This calculation was based on an equation describing the dissolution of a single particle under fairly restricted conditions (MERCER, 1967, eq. (3)). The particle size distribution was expressed as particle frequency data in narrow diameter increments (48 size intervals were used), and a summation of the undissolved mass in each size interval was made for several values of dissolution time.

The current designs of EW aerosol generators are capable of batch aerosol generation only, and further refinement is necessary before EW particles can be produced for continuous uses. Additionally, the composition and sizes of the EW particles examined to date tend to fall within relatively limited ranges. Several methods for improving the EW aerosol generator are presently under consideration.

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