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ABSTRACT

The diameters of potassium sulfate particles entrained in combustion gas were found to be between 0.1 and 1.0 μm. The particles had been formed when gas containing vaporized K_2SO_4 and less than 0.2 wt % ash particles was cooled at rates of about 700 K/s. The size and number of these particles agree quite well with those calculated by a computer model of particle formation by homogeneous condensation of vapor in rapidly cooling gas. This model predicts that submicron ash particles will be formed by homogeneous condensation in an MHD channel and that the seed-ash particles subsequently formed in the convective section of the steam plant will have diameters of less than one micrometer. This paper (1) describes the techniques being used to determine the sizes and loadings of seed-ash particles, (2) presents measurements of particles formed under a variety of conditions at the Argonne test facility, and (3) compares these measurements with calculations.

INTRODUCTION

Mathematical modeling has indicated that the largest mass fraction of the entrained seed-ash material leaving a coal-fired MHD power plant will be particles less than one micrometer in diameter [1]. This situation will not only have a major influence on the design of the gas clean-up system, but will also impact the arrangement of the convective sections of the steam generator, because small particle sizes affect the deposition of solids on the steam heater tubes.

At Argonne National Laboratory (ANL), the fouling of convective heat transfer surfaces by seed and coal ash is being studied to obtain important design information for MHD steam plants. These data are needed because the MHD operating conditions are considerably different from those of conventional coal-fired steam plants. The combustion gas composition and the characteristics of particles entering the MHD downstream components differ fundamentally from the gas and particles in the radiant furnace of a conventional coal-burning steam plant.

The size distribution of the entrained seed and slag particles in the MHD system will also be quite different from the sizes of the fly ash in conventional steam plants. In a conventional plant, the entrained particles are the ash inclusions in the coal, which typically have sizes of 10 μm or greater. In an MHD plant, all of the seed material along with some of the slag vaporizes in the combustor. Most of the slag

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vapor condenses homogeneously in the MHD channel to form submicron particles which are carried into the convective sections. The gas entering the MHD radiant boiler will contain numerous submicron liquid slag particles and a large quantity of vapor-phase potassium compounds, such as K, KO, KOH and K_2SO_4. As the combustion gas is cooled to the K_2SO_4 dew point, the KO and KOH are converted to K_2SO_4 in the oxidizing, sulfur-rich atmosphere downstream from the secondary combustor, and the sulfate begins to condense. Below the dew point, K_2SO_4 condenses either through nucleation to form new seed particles or on the already present slag particles. Eventually, these particles solidify. Thus, the gas in the MHD steam plant can contain condensible K_2SO_4 vapor or solid or liquid particles of ash and seed.

Estimates of particle sizes in coal-fired, open-cycle MHD plants have been made by Im and Ahluwalia, using models of ash and seed vapor condensation by both homogeneous and heterogeneous mechanisms [2,3]. Used in conjunction with models of particle growth and deposition, the entrained particle sizes and loadings, and the fouling rates in MHD steam bottoming plants can be estimated. These calculations show that, in a coal-fired MHD plant, 10 to 30% of the total coal ash can be vaporized in the combustor. Most of this vapor will condense homogeneously in the channel because of the rapid gas cooling there. This will result in the formation of particles in the 0.1 to 0.5 μm diameter range. Because the mechanisms causing these submicron particles to deposit in the radiant boiler are weak, a large fraction will be carried into the convective sections of the steam plant [4]. For the surface area of the slag particles and the gas cooling rates expected in a combined cycle MHD plant, the seed vapor will condense on the entrained, submicron ash particles as the gas temperature falls below the seed dew point ($\sim$1650 K).

Current investigations at Argonne National Laboratory are directed toward measuring seed-ash particle sizes and loadings, determining the deposition rates of these particles on cooled tube surfaces, and developing a computer code to calculate fouling rates. The objective is to improve and validate this code for use in the design of large, combined-cycle MHD power plants.

EXPERIMENTAL

The experiments to determine seed particle sizes were conducted on the oil-fired test leg of the Fossil Energy Users Laboratory (FEUL) at the Argonne National Laboratory. Figure 1 is a schematic of the test train, which is described in detail elsewhere [5]. For these tests, a mixture of heating oil, K_2SO_4, and fly ash was burned with air preheated to 800 K to produce about 0.5 kg/s of simulated MHD combustion gas. The experiments were carried out in an oxidizing atmosphere (stoichiometric ratio of 1.10) with a K/S atom ratio slightly less than 2.0 because of a small amount of sulfur present in the heating oil. An oxidizing atmosphere will be required for the convective section of an MHD steam plant to avoid severe corrosion of steam- and air-heater tubes.

The gas was cooled from the combustion temperature of 2300 to 2400 K to the 1200 to 1700 K range by flowing through a 6.5-m-long section of 0.56-m-ID water-cooled pipe. The gas then passed over a bank of vertical, air-cooled tubes simulating a steam or air heater. Similar experiments to determine seed-ash deposition rates and the effects of fouling deposits on heat transfer have been described previously [6,7]. In the experiments, K_2SO_4 in the oil was vaporized in the combustion zone, and as the gas cooled in the water-cooled sections, the seed vapor condensed by homogeneous nucleation to form a large number of submicron particles. Because of the low surface area of entrained ash particles and the high gas cooling rate, only a small fraction of the seed vapor condensed on the ash particles. The gas cooling rates in the FEUL test train were determined from the measured heat losses to the cooling water system.

To determine the number density and size distribution of seed particles, a specially designed sampling train has been installed in FEUL. A schematic of the sampling train is shown in Figure 2. The particles were sampled from the hot combustion gas in the middle of the seed condenser test section (see Figure 1). A combustion gas sample was withdrawn through a ceramic tube that extended several centimeters into the gas stream. A jet pump, an "air mover," driven by filtered, CO_2-free air, provided the pumping power and also served to dilute the gas sample by a factor of 20 to 60. The diluted gas flowed through a 9.8-cm-ID duct from which a sample was withdrawn isokinetically by a second air mover. This resulted in a second dilution by a factor of 15 to 25. Overall dilution factors of 300 to 1500 could thus be achieved. The large dilution factors were necessary in order to perform particle measurements reliably.

The twice-diluted gas sample flowed through a second 9.8-cm-ID tube from which gas-particle samples were withdrawn isokinetically into a variety of commercial instruments for particle counting and sizing. Proper isokinetic sampling was insured by maintaining laminar flow (Reynolds number less than 2000) in both tubes and by withdrawing the correct sample volume on the tube centerline.

The instruments used were (1) an electrical aerosol analyzer, EAA (TSI Model 3030), which measures and counts particles in the 0.01 to 1.0 μm diameter range, (2) an optical particle counter (Royco Model 225), which covers the 0.3 to 10 μm diameter range, (3) an electrostatic aerosol sampler, EAS (TSI Model 3100), which collects particles for electron microscopy, and (4) an impactor, which collects particles for chemical analysis. In operation, one or both of the air movers were adjusted until the dilution factor was sufficient that readings could be obtained with the aerosol analyzer and the optical counter. Samples of the twice-diluted gas had to be further diluted 100 times, using a Royco Diluter Model 252, for the optical particle counter to perform reliably.

The exact overall dilution factor was determined by on-line CO_2 analysis of the combustion gas and the twice-diluted gas sample. To determine large dilution factors, the CO_2 in the air supplied to the air movers was removed by adsorption on 13X Type molecular sieve, making it possible to measure accurately the concentration of CO_2 in the diluted samples without any interference. With knowledge of the initial CO_2 concentration in the test train, it was therefore possible to calculate the dilution factor. This factor, together with results obtained by the instruments, was then used in the determination of particle loading in the seed condenser test section.

The operating principle of the electrical aerosol analyzer (EAA) is based on measuring the distribution of electrical mobility of charged particles [8,9]. A gas sample is drawn into the instrument at 4 L/min and passes through a unipolar-ion diffusion charger. The charged aerosol gas sample then flows to the mobility analyzer. Particles with mobilities less than the cutoff mobility (determined by the voltages on the tubes) pass out of the analyzer and are collected in a high-efficiency filter. An electrometer continuously monitors the current produced by the capture of the charged particles in the filter. Since there is a monotonic relationship between mobility and particle size, the difference in current measured at two analyzer voltage settings is related to the number of particles in the size range defined by the cutoff size of the two voltage settings [8,9]. Experimental data are fed into an Apple II Plus microcomputer and stored on a disk for further detailed analysis.

In the optical particle counter, the gas sample flows through a focused light beam as a thin stream surrounded by sheath air so that only one particle at a time is illuminated and scatters light to the detector. The particle size distribution by count is obtained from the accumulated counts in each size channel. Measurement by this instrument is based on the assumption that the scattered light intensity is a monotonic function of particle size, but this is not always the case, since the response of optical particle counters depends not only on the size but also on the refractive index of the particles. For particles of unknown refractive index, as in our case, the error in particle size can range from 30 to 400%. For some refractive indices, the response curve may not necessarily be monotonic and more than one size of particles might correspond to a particular signal.

RESULTS AND DISCUSSION

Typical conditions of runs in which particle measurements were obtained are listed in Table I. In these tests, the K_2SO_4 vaporized at the combustor condensed homogeneously in the third gas conditioning section (see Figure 1) where the gas cooling rate was approximately 700 K/s. The particle samples were obtained from the transition section downstream from the tube bank at gas temperatures between 1400 and 1500 K. A representative normalized size distribution of the seed particles determined by the electrical aerosol analyzer is shown in Figure 3. These data have been corrected for an average dilution factor of 691. In other measurements, the average particle sizes ranged from 0.15 to 0.35 μm, a range similar to that shown in Figure 3.

Table I. Typical Experimental Test Conditions

Air Preheat Temperature	700-800 K
Combustion Temperature	2300-2400 K
Combustion Air Flow	0.45-0.46 kg/s
Fuel Flow[a]	0.037-0.038 kg/s
Stoichiometric Ratio[b]	1.10
Total Hot Gas Flow	0.49-0.50 kg/s
Cooling Rate During Nucleation	600-800 K/s
Test Duration	4-8 h
Gas Temperature near Sample Probe	1400-1500 K

[a] Fuel contained 25 wt % K_2SO_4 and 2.5% ash.

[b] Ratio of oxygen supplied to oxygen required for complete oxidation of oil.

Measurements obtained by the optical counter are not shown in Figure 3 because the results covered only the diameter range above 0.5 μm, where there were relatively few particles. Generally, the number of particles determined by the optical counter was a factor of four below the number measured by the EAA. This discrepancy may have been caused by the fact that the measurement errors for both instruments were greatest in the overlapping size range of 0.5 to 1.0 μm. In addition, the two instruments operate on different principles, as discussed in a previous section. Samples of particles were also obtained with the electrostatic aerosol sampler (EAS) and the impactor, but these samples have not been analyzed. Examinations of such samples obtained in other tests in FEUL [10] have shown the small particles to be K_2SO_4 and the large particles to be ash that is coated with a thin layer of a potassium salt, as one would expect.

The solids loading of the combustion gas calculated from the measured particle number densities was only approximately half of the solids known to be in the gas stream. The known quantity of gas-borne solids (about 1.8 wt % of the gas) was determined from the amount of seed and ash in the oil minus the measured amount of seed and ash deposited on the test train walls upstream from the sample point. The difference between the measured and known solids loadings was the result of material deposited on the walls of the ceramic sampling probe and in the first air mover. To overcome this problem, a new particulate sample probe is being assembled that will have porous walls purged with air to prevent particle deposition.

From the results obtained in the FEUL tests, it was found that in all cases where the gas temperature was low enough for large amounts of K_2SO_4 to condense in the test train, the seed particles had diameters between 0.1 and 0.6 µm. Calculations using the Im-Ahluwalia condensation code [3] indicated that in the FEUL test train, most of the K_2SO_4 vapor would condense homogeneously rather than on the ash particles. This was a result of (a) the high gas cooling rate ($\sim$700 K/s) in the test train section where seed condensed, and (b) the low specific surface area ($\sim$5 m^2/m^3) of condensation sites (the entrained ash particles). Calculated size distributions of K_2SO_4 particles formed in this manner are shown in Figure 4 for a gas cooling rate of 700 K/s and for two values of the parameter z_o which corrects the surface tension, $\sigma(r)$, of very small droplets for surface curvature in the equation

$$\sigma(r) = \frac{\sigma_o}{1 + (2z_o/r)} ,$$

where σ_o is the surface tension of bulk liquid. The parameter, z_o, has not been measured directly for K_2SO_4, but an estimate of 6 Å was derived from the measurements.

Calculations with the Im-Ahluwalia model have also shown that the particle size distribution and loading depends on the gas cooling rate during the condensation process. This is shown by the particle size distribution functions of Figure 5. The average size of particles formed at the 340 K/s gas cooling rate is nearly twice the size of particles formed at 700 K/s. At greater cooling rates, the saturation ratio of K_2SO_4 vapor reaches larger values (near 2) before nucleation begins. (The saturation ratio was defined as the ratio of the partial vapor pressure of K_2SO_4 to the equilibrium K_2SO_4 vapor pressure.) Thus, a larger amount of vapor nucleates, forming more of the initial particles, which have a diameter of only 26 Å. In addition, a greater cooling rate decreases the total amount of vapor originating from the more volatile, small particles. The vapor formed by this process contributes to the growth of the large particles at the expense of small particles by condensing on the less volatile, large particles.

CONCLUSIONS

The sizes and number densities of K_2SO_4 particles formed in a rapidly cooling stream of combustion gas have been determined in tests at Argonne's FEUL facilities. The average diameters after the nucleation is complete (between 0.15 and 0.35 µm) are in approximate agreement with those calculated by a computer code, which models particle formation by homogeneous nucleation. Thus, the experimental results strongly support the general validity of this model.

Particle sizes calculated for various conditions show that the average size is strongly dependent on the gas cooling rate during the condensation process; faster cooling leads to smaller particles. In addition, the size is a function of the physical parameter that corrects surface tension for the curvature of very small droplets. This parameter has not been measured directly for materials of interest to MHD, but an approximate value for K_2SO_4 was derived from the experiments. Presumably, values for liquid coal ash could be obtained from measurements of coal ash particles produced in MHD experiments.

When applied to the conditions of an open-cycle MHD plant, the particle formation code predicts that large numbers of submicron coal slag particles will form in the MHD channel by the same mechanism by which seed particles were formed in the FEUL experiments. This result has several important impacts on the design of the MHD steam bottoming plant. The presence of a large number of small ash particles will significantly increase radiant heat flux in the radiant boiler and the primary steam superheater [11]. Because their deposition rate is relatively low, most of the ash particles will be carried with the combustion gas into the convective sections of the steam generator. The particle formation code predicts that most of the seed vapor will condense on the ash particles [3] because of their high surface area, rather than condensing homogeneously as in the FEUL experiments. The resulting seed-ash particle sizes, however, will be similar to those of the seed particles formed in the FEUL experiments. These submicron particles tend to follow the flow streamlines, and thus turbulent and inertial impaction are relatively unimportant deposition mechanisms compared to thermophoresis. A large fraction of these small particles will be carried through the steam generator and must be removed from the stack gas by a highly efficient electrostatic precipitator or bag filter.

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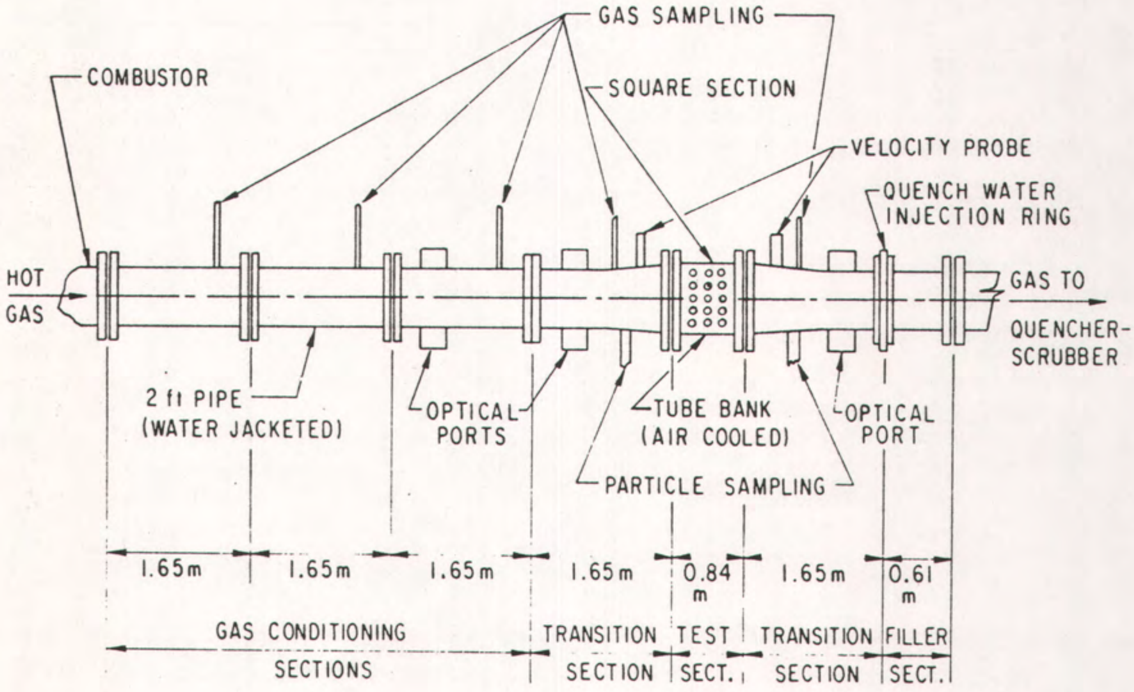

Figure 1. FEUL Test Train for Fouling Experiments.

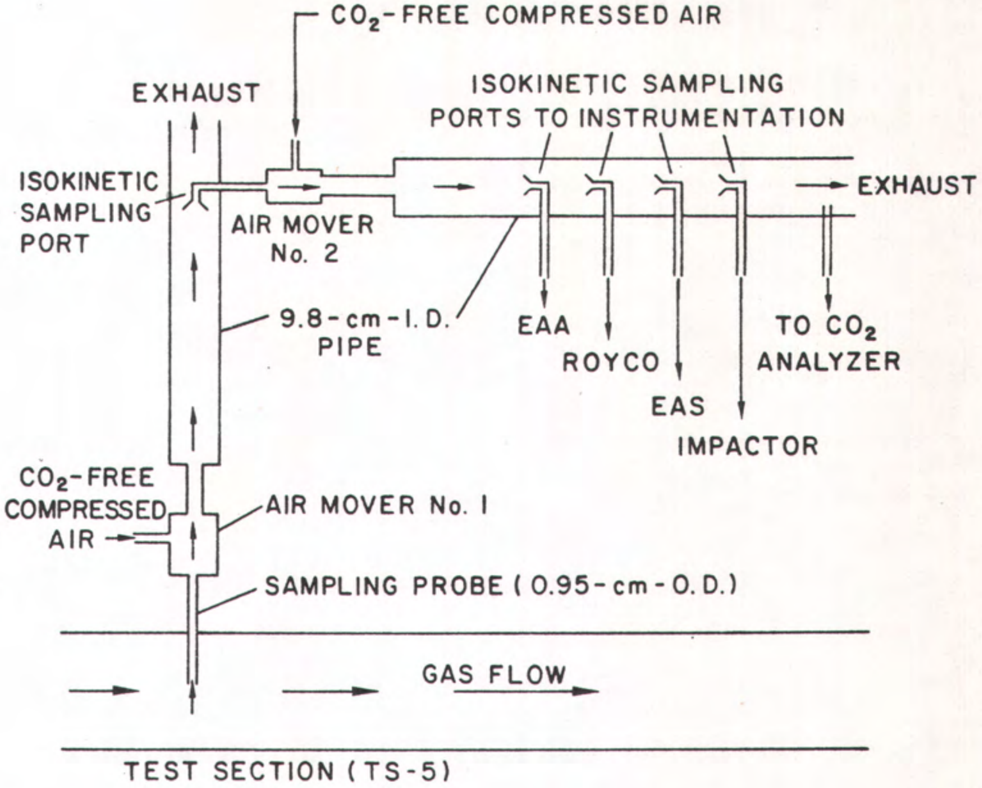

Figure 2. Sampling Test Train Used for Seed Particle Measurements.

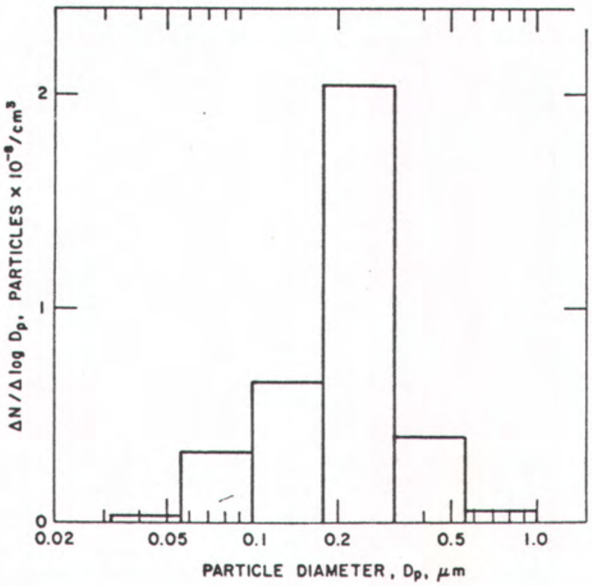

Figure 3. Normalized Size Distribution of Seed Particles Measured in FEUL Test Train.

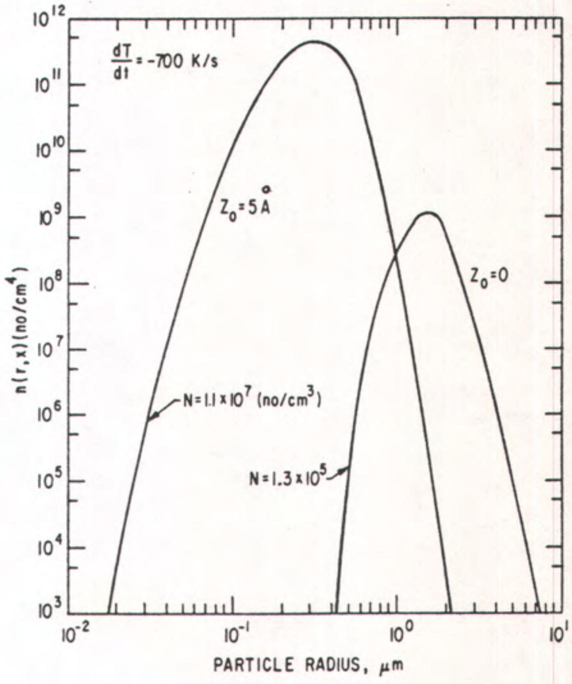

Figure 4. Seed Particle Size Distribution Functions for Two Different z_o Parameters.

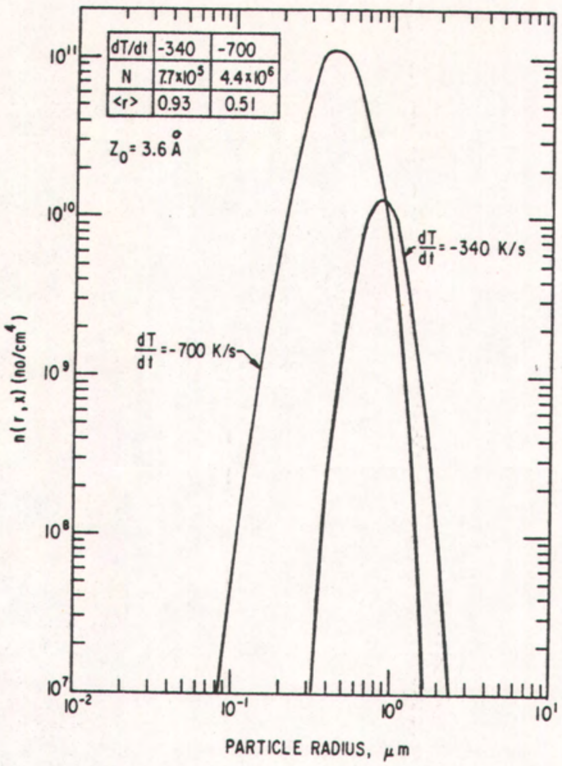

Figure 5. Effect of Different Gas Cooling Rates on Seed Particle Distribution Function.