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INTRODUCTION

The behavior of aerosols assumed to be characteristic of those generated during light water reactor (LWR) accident sequences and released into containment is being studied in the Nuclear Safety Pilot Plant (NSPP) which is located at the *Oak Ridge National Laboratory (ORNL)*. This project, which is part of the ORNL Aerosol Release and Transport (ART) Program, is sponsored by the Division of Accident Evaluation, Nuclear Regulatory Commission, and the purpose is to provide experimental qualification for LWR aerosol behavior codes under development.

The program plan for the NSPP aerosol project provides for the study of the behavior, within containment, of simulated LWR accident aerosols emanating from fuel, reactor core structural materials, and from concrete-molten core materials interactions. The aerodynamic behavior of each of these aerosols was studied individually to establish its characteristics; current experiments involve mixtures of these aerosols to establish their interaction and collective behavior within containment. Tests have been conducted with U_3O_8 aerosols, Fe_2O_3 aerosols, and concrete aerosols in an environment of either dry air [relative humidity (RH) less than 20%] or steam-air [relative humidity (RH) approximately 100%] with aerosol mass concentration being the primary experimental variable. Experiments are underway involving mixtures of these aerosols, and, to date, the test aerosol mixture has been Fe_2O_3 + U_3O_8; in these tests the primary experimental variables have been aerosol mass concentration and aerosol mass ratio.

EXPERIMENTAL

The NSPP facility, shown schematically in Fig. 1, includes a test containment vessel, aerosol generating equipment, analytical sampling and system parameter measuring equipment, and an in-vessel liquid spray decontamination system. The NSPP vessel is a stainless steel cylinder with dished ends having a diameter of 3 m, a total height of 5.5 m, and a volume of 38.3 m^3. The floor area is 7.7 m^2 and the internal surface area (including top, bottom, and structural items) is 68.9 m^2. The equipment for the measurement of aerosol parameters includes filter samplers for measuring the aerosol mass concentration, coupon samplers for aerosol fallout and plateout measurement, cascade impactors and a centrifuge sampler for determining the aerodynamic particle size distribution of the aerosol, and devices for collecting samples for electron microscopy. System parameters measured are moisture content of the vessel atmosphere, steam condensation rates on the vessel wall, temperature of vessel atmosphere, temperature gradients near the wall, and vessel pressure.

For the dry aerosol tests the vessel atmosphere was dry air (RH <20%) and the temperature and pressure were slightly above ambient. The slight elevations in temperature and pressure result from the heat produced and gases injected by the plasma torch aerosol generator.

The steam-air aerosol tests were conducted under quasi-steady-state steam conditions. The test atmosphere was prepared by injecting steam into the vessel (initially at subatmospheric pressure) to form a steam-air mixture at elevated temperature and pressure (around 380 K and at an absolute pressure of about 0.2 MPa); upon achieving this condition the rate of steam injection was reduced and the accumulated steam condensate removed from the vessel. The test aerosol was then introduced and steam injection was continued for six hours at a low rate to balance steam losses by wall condensation and assure maintenance of the quasi-steady-state conditions.

Single-Component Aerosol Tests

A number of single-component aerosol tests have been conducted under both dry air and steam-air test environments. Table I lists these tests; results from these tests have been reported [1, 2, 3].

Under dry conditions the three aerosols, U_3O_8, Fe_2O_3, and concrete, behave in a different manner with regard to rate of removal (decrease in aerosol mass concentration) from the vessel atmosphere. Figure 2 compares the behavior of the three aerosols. (Note that the aerosol mass concentration is normalized with respect to maximum concentration and that time is measured from the time of termination of aerosol generation for the purpose of comparison.) Scanning electron microphotography (SEM) shows the aerosols to be agglomerated in the form of branched-chains (Fig. 3). Particle size measurements by cascade impactors and spiral centrifuges indicated that the aerodynamic mass median diameter (AMMD) [4] of the U_3O_8 and Fe_2O_3 aerosols ranged between 1.5 and 3 μm while that of the concrete aerosol was about 1 μm, or less. Based upon the results from these tests under dry conditions, it has been observed that these aerosols have similar sizes and shapes but act aerodynamically in a different fashion.

The presence of steam in the test environment causes a change in both the aerodynamic behavior and the physical shape of these aerosols. The aerodynamic behavior of the aerosols is compared in Fig. 4. The most obvious effect of steam is an enhanced rate of aerosol removal from the vessel atmosphere in the case of U_3O_8 and Fe_2O_3 aerosols. For example, in Fig. 2 under dry conditions, the time required for 99% of the Fe_2O_3 aerosol to disappear from the vessel atmosphere is about 350 min.; under steam-air conditions this time is about 100 min. A similar comparison can be made for U_3O_8 aerosol. The shape of these two aerosols is changed from chain-agglomerate to almost spherical by the presence of steam as illustrated in Fig. 5 for U_3O_8. The AMMD for the U_3O_8 or Fe_2O_3 aerosols in steam range from about 1 to 2 µm.

Concrete aerosol does not seem to be affected by the presence of steam in the same manner as U_3O_8 or Fe_2O_3 aerosol. This lack of influence is illustrated in Fig. 6 where the rates of removal of concrete aerosol under dry and under steam-air conditions are compared. This aerosol was generated by passing powdered limestone-aggregate concrete through the plasma torch aerosol generator. The concrete aerosol is not a simple, single-component, aerosol such as U_3O_8 or Fe_2O_3; it is actually a complex mixture of Al_2O_3, SiO_2, CaO, MgO, Fe_2O_3, and various silicates with Al, Ca, Mg, and Fe as the cations. Steam also affects the physical shape of concrete aerosols (possibly to a slightly lesser degree than for U_3O_8 or Fe_2O_3) producing some spherical agglomerates. Figure 7 contains scanning electron microphotographs of a concrete aerosol in a dry air and in a steam-air atmosphere.

Multi-Component Aerosol Tests

Recent activities in the NSPP involve the study of the behavior of multi-component (mixed) aerosols in both dry air and steam-air environments. Details of these tests are contained in Table II. The first mixed aerosol to be studied in detail is $U_3O_8 + Fe_2O_3$. This mixture simulates those aerosols emanating from molten fuel and molten-core support and structural materials. Experimental procedures are essentially the same as for the single-component aerosol tests. The principal difference is in aerosol generation; the U_3O_8 and Fe_2O_3 aerosols are produced with separate plasma torch generators and allowed to mix within the vessel.

Four mixed aerosol experiments involving various mixtures of Fe_2O_3 and U_3O_8 aerosols have been completed; three were conducted in a steam-air environment and one in a dry air (RH <20%) environment. The behavior of the mixed aerosol ($Fe_2O_3 + U_3O_8$) in a steam-air environment has been similar in the three experiments conducted, although the mass ratio of F_2O_3 to U_3O_8 has been different in each case. The aerosol mass fraction airborne (C/C_{max}) as a function of time after termination of aerosol generation is illustrated in Fig. 8 for these experiments. Although the rate of aerosol removal during the first 30 min is somewhat larger in Exps. 611 and 613 as compared to Exp. 612, the time required for 99% removal of aerosol mass from the volume of the vessel is about 60 min in all three experiments. SEM photographs of the mixed aerosol showed almost spherical clumps of aerosol in each case. The AMMD of the mixed aerosol in all cases was in the 1 to 1.7-µm range.

To illustrate the effect of steam on the behavior of the mixed aerosol, the results from experiment 631 are compared with those of Nos. 611-613 in Fig. 8. Under dry air conditions, the mixed aerosol tends to remain airborne longer than under steam-air conditions. Note that the time required for 99% of this aerosol to be removed from the vessel is about 400 min as compared with 60 min for the aerosol in the steam-air environment. SEM photographs show the aerosol to be in the form of chain-agglomerates (also observed in previous experiments with Fe_2O_3 or U_3O_8 aerosol in dry air) rather than in spherical clumps as in Nos. 611-613. The AMMD for the mixed aerosol is slightly larger in the dry atmosphere with a value as large as 2.7 μm being observed.

It appears, based upon data in hand, that the influence of one aerosol component on the other, in a mixed aerosol, can be significant. The behavior of a mixed Fe_2O_3 + U_3O_8 aerosol is compared with that of a Fe_2O_3 aerosol and a U_3O_8 aerosol in Fig. 9. The behavior (rate of aerosol mass removal) of the Fe_2O_3 + U_3O_8 aerosol is more similar to that of Fe_2O_3 aerosol that that of U_3O_8 aerosol. Future tests on mixed aerosols will permit a more definitive examination of the influence of one component on another in mixed aerosols.

SUMMARY

General statements may be made on the behavior of single-component and multi-component aerosols in the NSPP vessel. The removal processes for U_3O_8, Fe_2O_3, and U_3O_8 + Fe_2O_3 aerosols are enhanced in a steam-air atmosphere. Steam-air seems to have little effect on removal of concrete aerosol from the vessel atmosphere. A steam-air environment causes a change in aerosol shape from chain-agglomerate to basically spherical for U_3O_8, Fe_2O_3, and U_3O_8 + Fe_2O_3 aerosol; for concrete the change in aerosol shape is from chain-agglomerate to partially spherical. The mass ratio of the individual components of a multi-component aerosol seems to have an observable influence on the resultant behavior of these aerosols in steam.

The enhanced rate of removal of the U_3O_8, the Fe_2O_3, and the mixed U_3O_8 + Fe_2O_3 aerosols from the atmosphere of the NSPP vessel by steam-air is probably caused by the change in aerosol shape and the condensation of steam on the aerosol surfaces combining to increase the effect of gravitational settling. The apparent lack of an effect by steam-air on the removal rate of concrete aerosol could result from a differing physical/chemical response of the surfaces of this aerosol to condensing steam.

REFERENCES

1. Adams, R. E., et al., "Influence of Steam on the Behavior of U_3O_8 Aerosols," Proceedings of the USNRC Tenth Water Reactor Safety Research Information Meeting, Gaithersburg, MD, October 12—15, 1982, NUREG/CP-0041, Vol. 2 (January 1983).

2. Adams, R. E., "Behavior of U_3O_8, Fe_2O_3, and Concrete Aerosols in a Condensing Steam Environment," Proceedings of the USNRC Eleventh Water Reactor Safety Research Information Meeting, Gaithersburg, MD, October 24—28, 1983, NUREG/CP-0048, Vol. 3 (January 1984).

3. Quarterly Aerosol Release and Transport Program Progress Reports for the years 1980—1984. R. E. Adams and M. L. Tobias, editors.

4. Mercer, T.T., "Aerosol Technology in Hazard Evaluation,," New York, Academic Press (1973).

Table I. Details of single-component aerosol tests

Test Nos.	Aerosol	No. of tests	Test environment	Aerosol conc. range (μg/cm^3)
201-7, 209	U_3O_8	8	Air (dry)	0.05-9.0
208, 210	U_3O_8	2	Air (moist)	7.1, 12.5
401-4, 406-7	U_3O_8	6	Air-steam	5.8-28.0
511	Fe_2O_3	1	Air (dry)	2.4
501-5	Fe_2O_3	5	Air-steam	1.0-8.5
531	Concrete	1	Air (dry)	1.5
521-2	Concrete	2	Air-steam	1.1, 1.5

Table II. Details of multi-component aerosol tests

Test No.	Mixed aerosol	Test environment	Max. aerosol conc. (μg/cm^3)	Mass ratio (Fe_2O_3/U_3O_8)
611	U_3O_8 + Fe_2O_3	Air-steam	4.0 5.5	1.4/1
612	U_3O_8 + Fe_2O_3	Air-steam	1.8 0.5	0.3/1
613	U_3O_8 + Fe_2O_3	Air-steam	0.7 6.8	9.7/1
631	U_3O_8 + Fe_2O_3	Air (dry)	1.7 1.2	0.7/1

NSPP FACILITY SCHEMATIC

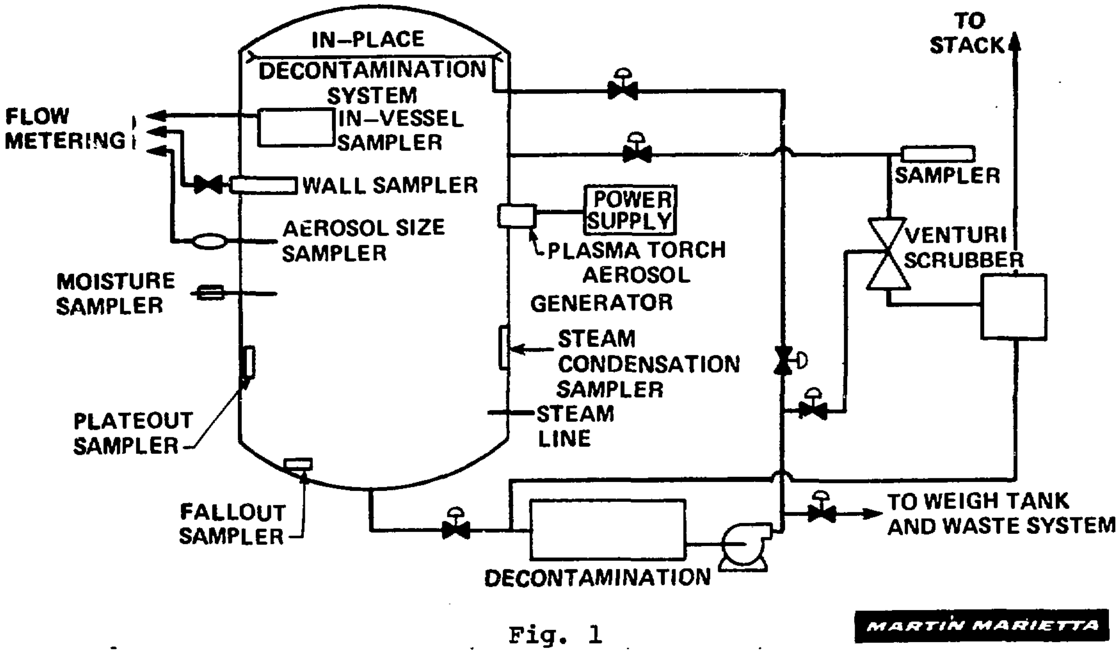


Fig. 1

BEHAVIOR OF VARIOUS AEROSOLS IN A DRY ENVIRONMENT (RH <20%)

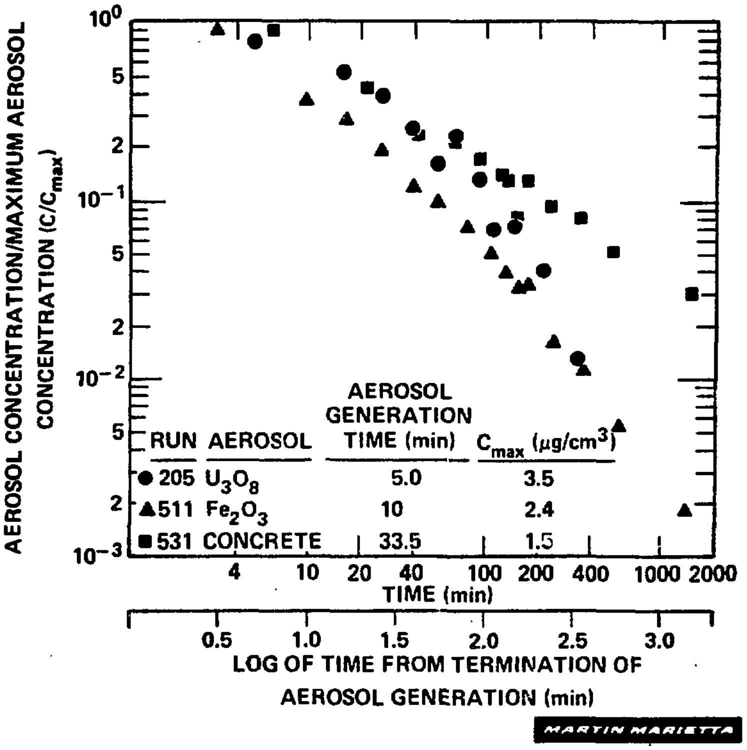
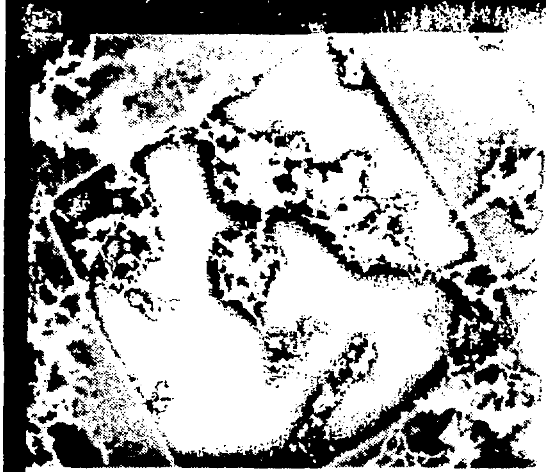


Fig. 2

TYPICAL CHAIN AGGLOMERATE AEROSOLS IN A DRY AIR ENVIRONMENT (RH < 20%)



TEST 207, U_3O_8, 2000X

TEST 511, Fe_2O_3, 2000X

TEST 531, CONCRETE, 2700X

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Fig. 3

BEHAVIOR OF VARIOUS AEROSOLS IN A STEAM–AIR ENVIRONMENT (RH ~ 100%)

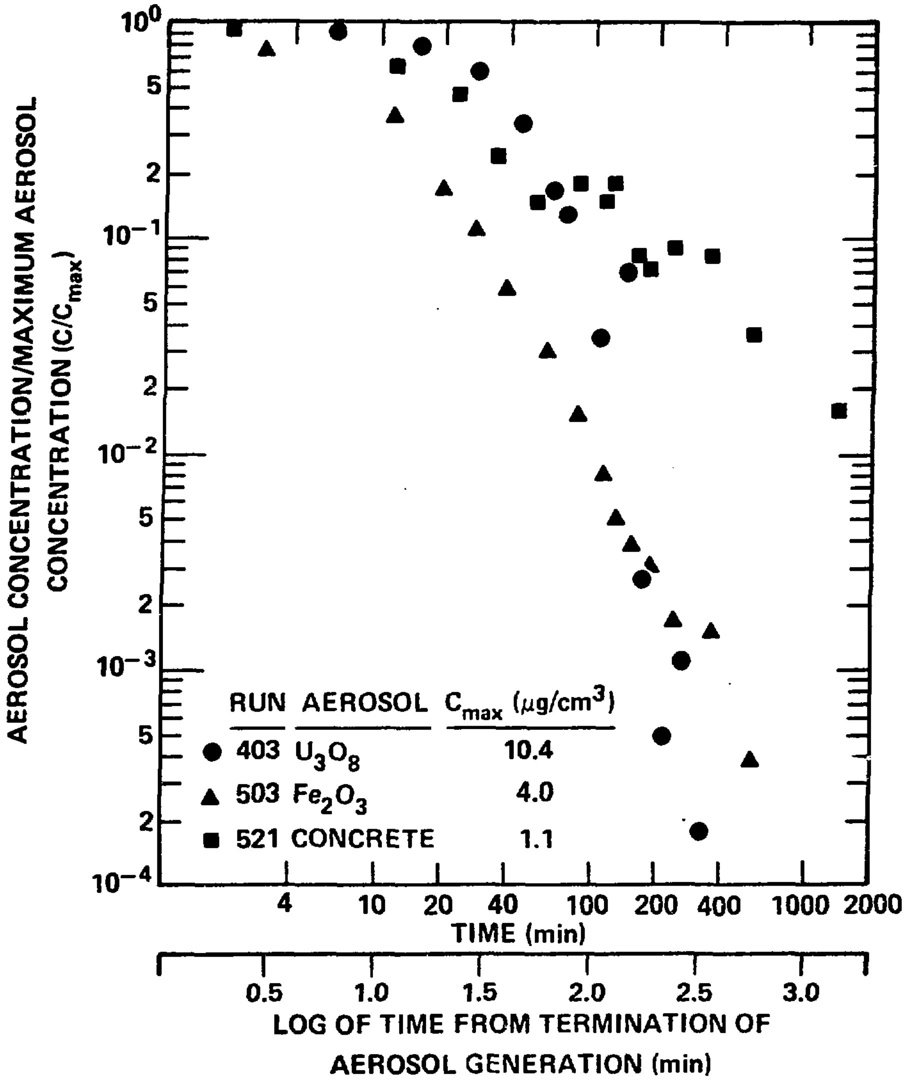

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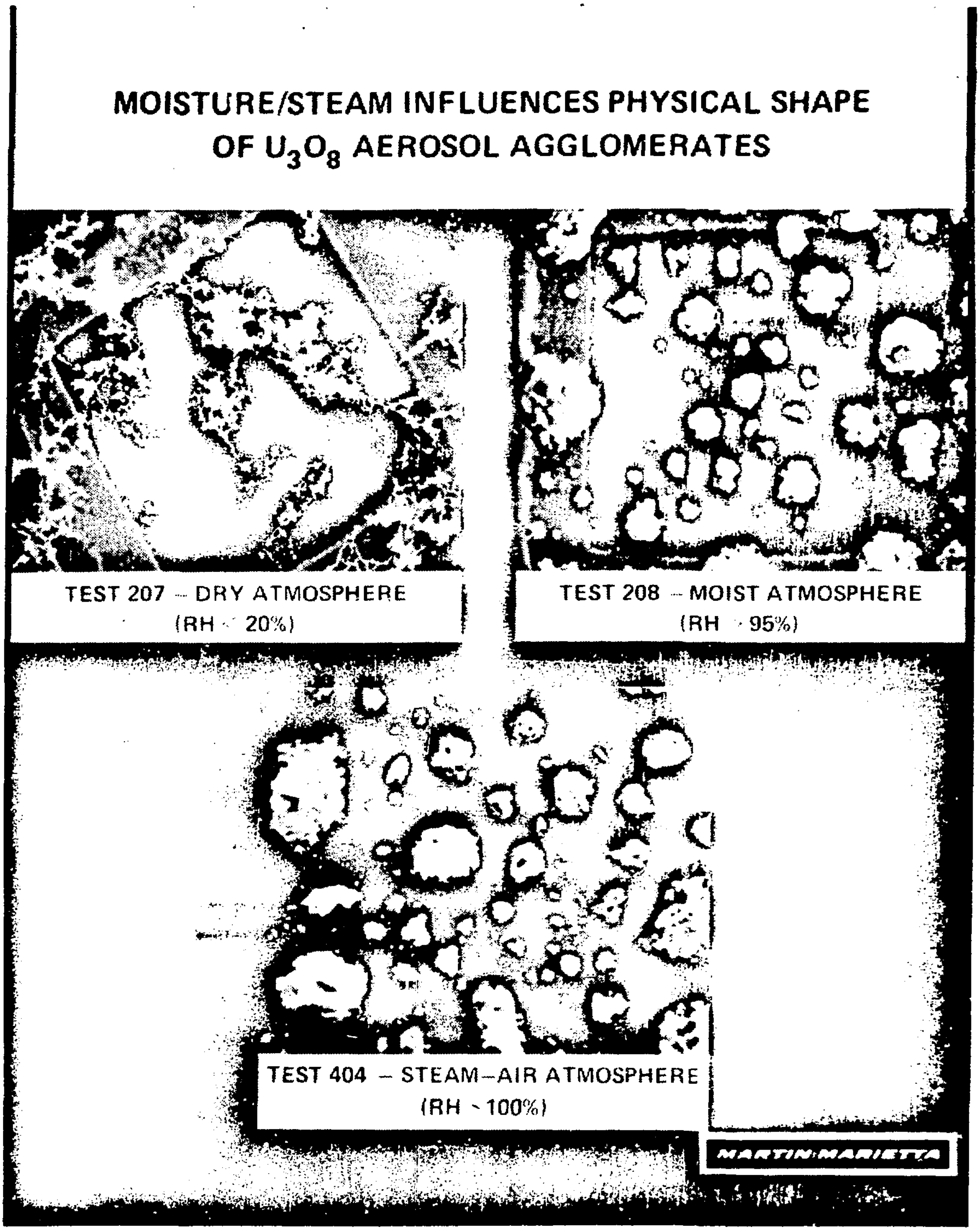
Fig. 4



MOISTURE/STEAM INFLUENCES PHYSICAL SHAPE OF U_3O_8 AEROSOL AGGLOMERATES

TEST 207 – DRY ATMOSPHERE
(RH $<$ 20%)

TEST 208 – MOIST ATMOSPHERE
(RH $>$ 95%)

TEST 404 – STEAM–AIR ATMOSPHERE
(RH ~100%)

Fig. 5

INFLUENCE OF STEAM ON BEHAVIOR OF CONCRETE AEROSOLS IN NSPP VESSEL

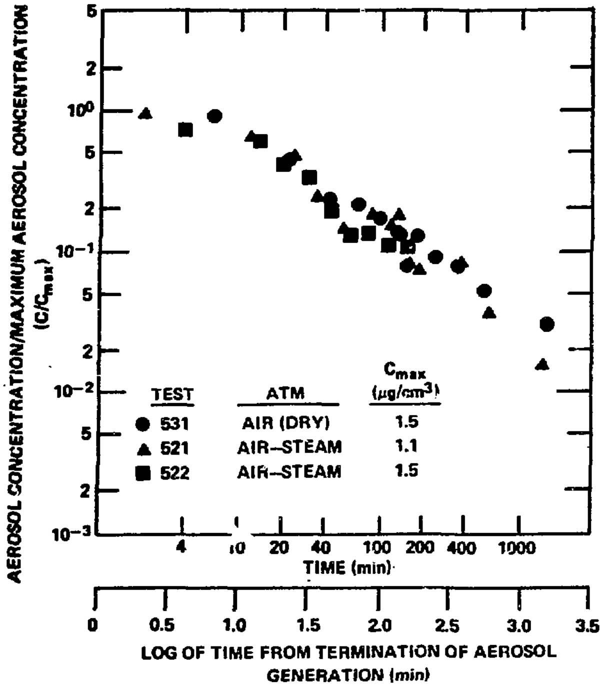


Fig. 6

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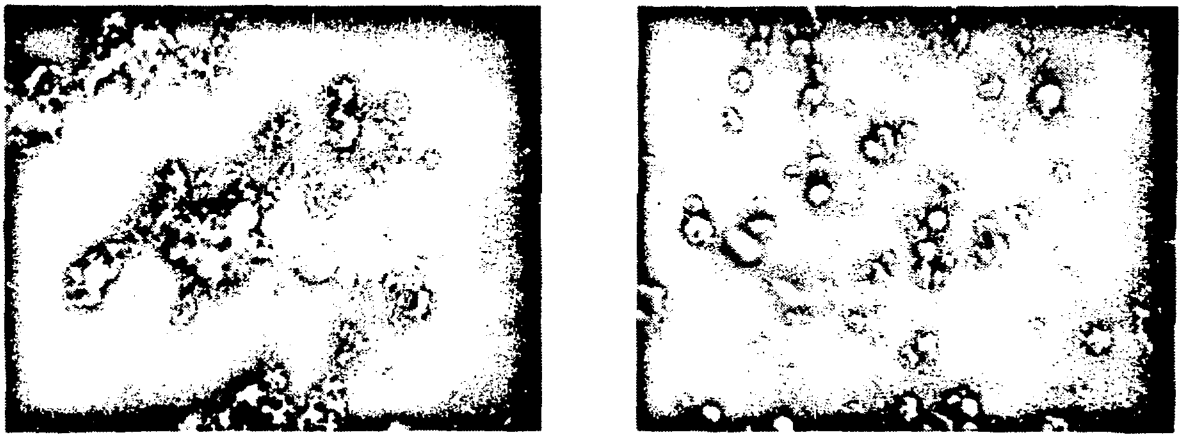
STEAM INFLUENCES PHYSICAL SHAPE OF CONCRETE AEROSOL AGGLOMERATES



TEST 531 – DRY ATMOSPHERE
(RH < 20%)
6300X MAG.

TEST 052 – STEAM–AIR ATMOSPHERE
(RH ~ 100%)
9000X MAG.

Fig. 7

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COMPARISON OF BEHAVIOR OF MIXED AEROSOLS (Fe_2O_3 + U_3O_8) IN STEAM–AIR AND DRY AIR ENVIRONMENTS

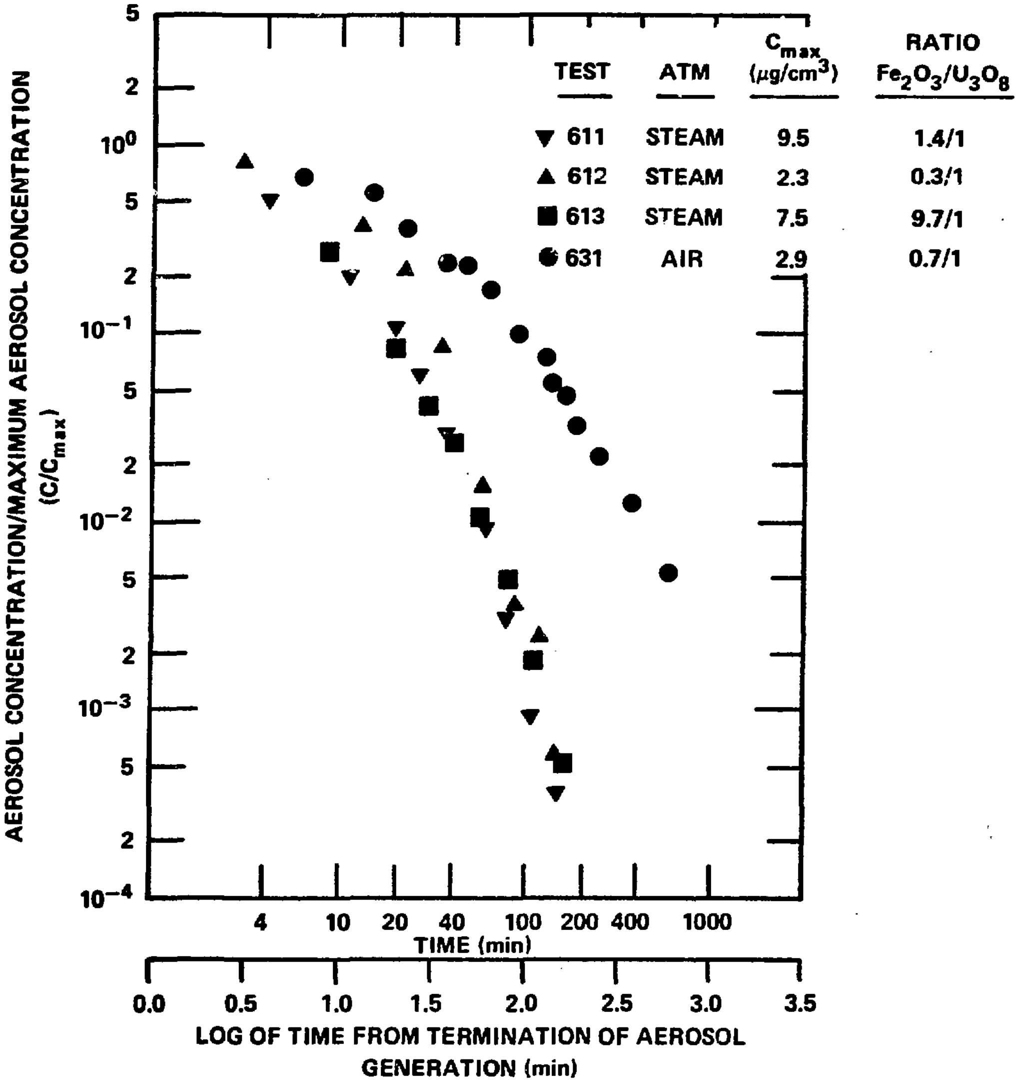

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Fig. 8

COMPARISON OF THE BEHAVIOR OF A MIXED–AEROSOL WITH THAT OF INDIVIDUAL AEROSOLS IN A STEAM–AIR ENVIRONMENT

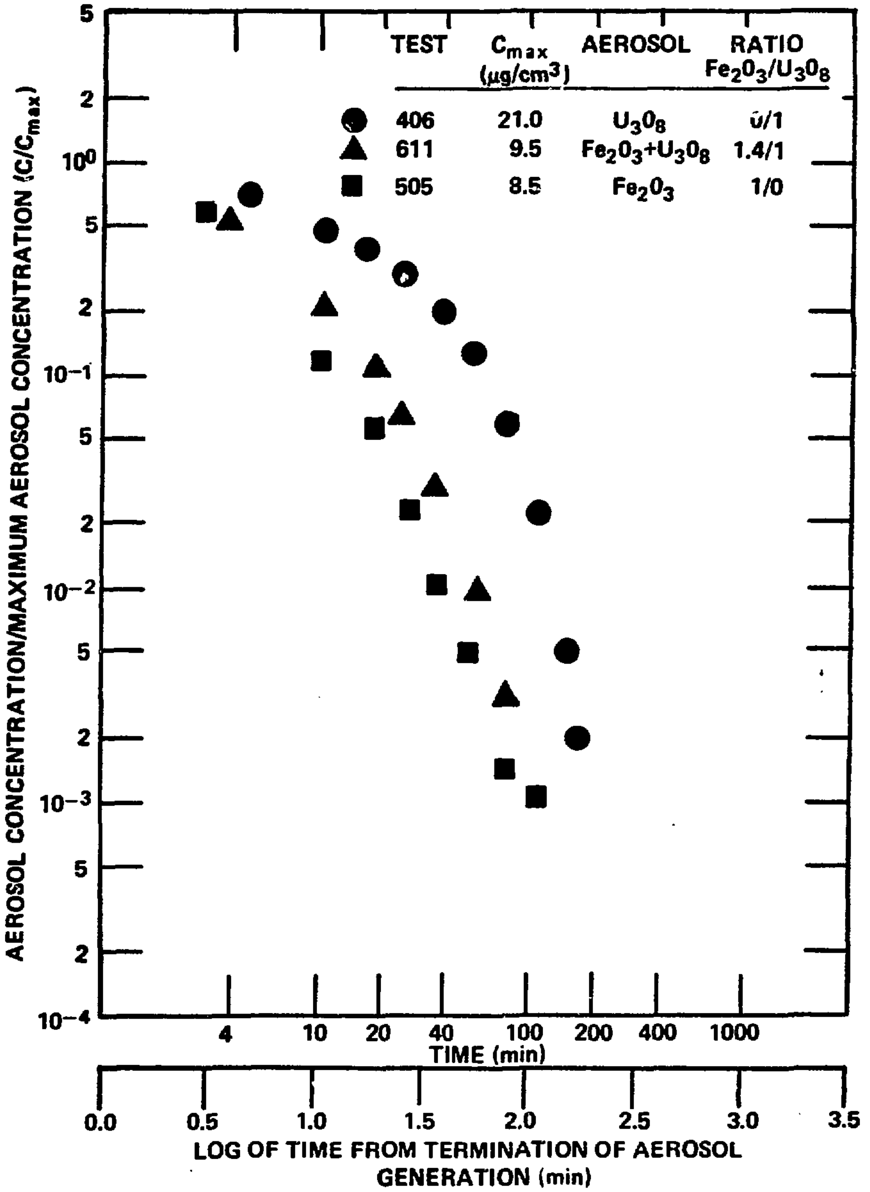

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Fig. 9