BEHAVIOR OF SODIUM OXIDE, URANIUM OXIDE AND MIXED SODIUM OXIDE-URANIUM OXIDE AEROSOLS IN A LARGE VESSEL

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ABSTRACT

Results are reported for experiments on the behavior of sodium oxide aerosols, uranium oxide aerosols, and various mixtures of these in the Nuclear Safety Pilot Plant (NSPP) vessel at Oak Ridge National Laboratory. The NSPP program, sponsored by the Nuclear Regulatory Commission, is to provide experimental qualification of the HAARM-3 aerosol behavioral code being independently developed at Battelle-Columbus Laboratories. The results of fourteen single-component and six mixed-component aerosol experiments are presented along with selected comparisons with code predictions.

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INTRODUCTION

A program is being conducted at the Oak Ridge National Laboratory (ORNL) to study the behavior of sodium oxide aerosols, uranium oxide aerosols, and mixtures of these in the 38.3-m³ containment vessel of the Nuclear Safety Pilot Plant (NSPP). The purpose of the program, sponsored by the Nuclear Regulatory Commission (NRC), is to provide the experimental qualification to the HAARM-3 aerosol behavioral code,[1] which is under independent development for NRC at Battelle-Columbus Laboratories (BCL).

Experiments are conducted to explore the behavior of various mixtures of the two major liquid-metal fast breeder reactor (LMFBR) nuclear aerosol species under conditions appropriate for speculated releases into the secondary containment caused by hypothetical LMFBR accidents. The program plan provides for studies of single-component aerosols of both sodium and uranium oxides to establish their individual characteristics and for studies to evaluate the behavior of mixtures of these aerosols at varying mass ratios and sequences of generation.

The NSPP facility 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 3.05 m, a total height of 5.49 m, and a volume of 38.3 m³. The floor area is 7.7 m², and the internal surface area (including top, bottom, and structural items) is 68.9 m². The equipment for the measurement of aerosol parameters includes filter samplers for measuring the aerosol mass concentrations, coupon samplers for aerosol fallout and plateout measurements, cascade impactors for determining the aerodynamic particle size distributions of the aerosol, and electrostatic precipitators for collecting samples for electron microscopy. Other system parameters measured are vessel atmosphere temperatures, wall temperature gradients, and vessel pressure.

EXPERIMENTAL

Sodium Oxide Aerosol Experiments

Fourteen single-component aerosol experiments have been conducted. In four of these experiments, sodium oxide aerosol was produced by sodium pool fires of 1, 5, 5, and 10 kg of heated sodium with maximum observed aerosol concentrations of 6, 11, 11, and 25 g/m³, respectively. (The two runs at 5 kg illustrate reproducibility of the data.) The observed mass concentration transients for these are given in Fig. 1. Aerodynamic diameters of the agglomerating aerosols increased from the source size up to as large as 4 μ. The burning sodium produced temperatures in the vessel atmosphere ranging from 323 to 358 K and pressures ranging from 0.110 to 0.112 MPa.

Three sodium oxide aerosol experiments were performed by injecting 5 or 10 kg of heated sodium into the vessel through a single spray nozzle. Estimated maximum aerosol concentrations at the time of termination of the sodium spray were in the 40- to 60-g/m^3 range. The aerodynamic mass mean diameter of the agglomerating aerosols increased rapidly from the source size to as large as ~7 μ and then decreased over 10 hr to about 1.5 to 2.0 μ. The fractional disappearance of this aerosol from the vessel atmosphere was consistent with those in the sodium pool fire experiments. The vessel atmosphere temperatures, as expected, were larger than those produced by sodium pool fires, and a gradient was produced from the bottom to the top of the vessel; maximum temperatures near the top of the vessel ranged from 473 to 673 K. The absolute pressures produced in the vessel ranged from 0.139 to 0.145 MPa. Both the temperature and pressure transients were of short duration, however.

Uranium Oxide Aerosol Experiments

Seven single-component uranium oxide aerosol experiments were also conducted. In three of these experiments, the aerosol was generated by vaporizing a uranium metal electrode in a dc-arc and allowing it to combine with the oxygen in the vessel atmosphere.[2] Four additional experiments were conducted using an aerosol generator adapted from a commercial plasma metallizing torch[3] in which powdered uranium metal and oxygen gas are introduced into an argon plasma by means of a special adapter/combustion chamber. Figure 2 presents the measured mass concentration transient for all seven of these tests.

When using the dc-arc generator, the duration of generation was long (compared with that in the sodium oxide experiments), and the maximum aerosol concentrations were small. For the three experiments, maximum aerosol concentrations of only 0.05, 0.13, and 0.22 μg/cm^3 were produced. The aerodynamic mass mean diameter of the agglomerating aerosol grew to a maximum of 1.8 μ. Temperature and pressure changes in the vessel atmosphere were small.

Larger uranium oxide aerosol concentrations were achieved with the plasma torch aerosol generator. The four experiments reported produced maximum aerosol concentrations ranging from 0.7 to 5. μg/cm^3 at termination of generation. At these higher concentrations, the fractional disappearance of the aerosol was larger than at the lower concentrations, and the aerodynamic mass mean diameter of the agglomerating aerosol became larger. During one of the larger concentration experiments, (run 205), agglomerated particle diameters in the 4- to 5-μ range were measured.

Mixed Sodium Oxide-Uranium Oxide Aerosol Experiments

A series of experiments is underway to investigate the aerodynamic
behavior of an admixture of sodium oxide and uranium oxide aerosols. It
is of interest to study the influence of each aerosol on the other, and
determine if the two aerosols act independently in a closed vessel, or
if the two aerosols mix and act as a "new" single-component aerosol with
properties different from those of the individual aerosol components.
This series of experiments was designed to investigate (1) the influence
of particle size on the agglomeration process wherein one aerosol is gen-
erated first, allowed to increase from source-size to larger sizes by ag-
glomeration, then mixed with a freshly-generated aerosol of source-size
particles, and (2) the influence of relative mass ratio of the two aerosols
wherein the two are generated and mixed simultaneously and allowed to ag-
glomerate from source-size particles. The experiments performed to date
are listed in Table I, below.

Table I
Mixed Sodium Oxide-Uranium Oxide Aerosol Experiments

Experiment (Run) No.	Sequence of Mixing	Mass Ratio at Mixing (Na_2O/U_3O_8)
301	Uranium Oxide First Followed by Sodium Oxide at 102 min	30/1
302	Uranium Oxide First Followed by Sodium Oxide at 93 min	4/1
306	Sodium Oxide First Followed by Uranium Oxide at 44 min	1/1
303	Simultaneous Generation	1/1.2
304	Simultaneous Generation	1/10
305	Simultaneous Generation	3.5/1

In all of these experiments, the initial atmosphere was air at a relative
humidity of less than 20%. Temperature and pressure generated in the ves-
sel by the experiments were similar to those produced by comparable single-
component sodium pool fire experiments.

Influence of aerosol size - The first mixed-oxide aerosol experiment (No.
301) was performed by generating uranium oxide (with the DC-Arc generator)
for a 20 min period, allowing a period of 82 min for agglomeration to pro-
ceed, and then introducing sodium oxide aerosol by means of a pool fire of

3 min duration. Figure 3 displays the results from this experiment. The uranium oxide aerosol concentration decreased over the 82 min period of agglomeration, until, at the time of initiation of the sodium pool fire, the concentration was an estimated 0.1 µg/cm^3. Upon introduction of the sodium oxide aerosol, the concentration of uranium oxide dropped significantly, and the rate of removal from the vessel atmosphere was similar to that of the sodium oxide component. This behavior, supported by the fallout data, suggested that the two aerosols were co-agglomerating and, for the most part, acting as a single aerosol. Figure 3 also contains, for comparison, the results from single component aerosol experiments with sodium oxide or uranium oxide. Run 203 illustrates the behavior that would be expected from a comparable single-component uranium oxide aerosol experiment; Run 101 illustrates the behavior of a comparable sodium oxide aerosol experiment. It is observed that the newly-generated sodium oxide aerosol has a large influence on the aerodynamic behavior of an agglomerated uranium oxide aerosol.

A similar experiment (No. 302), performed at a different aerosol mass ratio, produced results comparable to those observed in Run 301.

The influence of a newly-generated uranium oxide aerosol on an agglomerated sodium oxide aerosol was studied in Experiment (Run) 306. The sodium oxide aerosol was produced by a sodium pool fire of 26 min duration and then allowed to agglomerate for an additional period of 18 min, at which time the concentration was estimated to be 4.5 µg/cm^3. Uranium oxide aerosol generation (by the plasma torch generator) was initiated at the end of the agglomeration period and continued for 13 min. The results of this experiment are illustrated in Fig. 4. Upon introduction of the uranium oxide aerosol, the concentration of the sodium oxide aerosol decreased significantly. Over the period of 16 min after start of the uranium oxide aerosol generation, the sodium oxide aerosol concentration dropped about 92%. This behavior, supported by the fallout data, suggested that the two aerosols were co-agglomerating. Figure 4 also contains, for comparison, the results from single-component experiments with sodium oxide or uranium oxide aerosols; Run 104 illustrates the behavior that would be expected from a comparable sodium oxide experiment.

Influence of aerosol mass ratio - Three experiments have been conducted to investigate the influence of the mass ratio of the two components of a mixed-oxide aerosol on its aerodynamic behavior. In the first experiment, Run 303, the target mass ratio of the two aerosols was 1 to 1. Uranium oxide aerosol generation (with the plasma torch generator) was initiated first and followed by sodium oxide aerosol generation (by a sodium pool fire) 1.5 min later. The uranium oxide aerosol generation period was about 25 min and the sodium oxide aerosol generation period was about 12 min. Measured aerosol mass concentration for each component of this mixed-oxide aerosol as a function of time are given in Fig. 5. Over the first nine hours of the run, the measured mass ratio of uranium oxide to sodium oxide ranged over the interval from 1.2 to 2.5. The concentration values for sodium oxide, at times greater than 500 min, are probably slightly greater than actual values; the reagents used in dissolving the mixed-oxide samples were found to contain a low-level sodium impurity, and large uncertainty is associated with the corrections made to the measured values to compensate for this. The observed behavior of this mixed-oxide aerosol, supported by fallout data and other measurements, suggests that the two aerosols were co-agglomerating and the composite behavior was somewhat

different from that of a single-component aerosol. At the termination of the experiment at 119 hrs, the approximate distribution of the aerosol components was indicated to be 88% of the sodium oxide and 82% of the uranium oxide on the floor of the vessel and 12% of the sodium oxide and 18% of the uranium oxide plated onto internal surfaces.

The second experiment (Run 304) was to investigate the behavior of a mixed-oxide aerosol having a mass ratio of about 10 to 1 (uranium oxide to sodium oxide). Sodium oxide generation was initiated first and then followed by the start of uranium oxide aerosol generation 2 min later. The sodium oxide generation period was 3 min and the uranium oxide aerosol generation period was 19 min. At the termination of the uranium oxide aerosol generation period, the mass ratio of uranium oxide to sodium oxide was about 10 to 1. Over the first 8 hrs, the measured mass ratio decreased from about 10 to 1 to about 1 to 1. The behavior of this mixed-oxide aerosol was very similar to that produced in Run 303, with all measurements indicating co-agglomeration. The test was terminated at 48 hrs and the aerosol distribution indicated that 73% of the uranium oxide and 50% of the sodium oxide had deposited on the floor, and 27% of the uranium oxide and 50% of the sodium oxide had deposited on the internal surfaces of the vessel.

The third experiment (Run 305) was to investigate the behavior of a mixed-oxide aerosol at a 10 to 1 mass ratio of sodium oxide to uranium oxide. The uranium oxide aerosol was generated for 5.7 min and 1 min later, the sodium oxide aerosol generation was initiated by a sodium pool fire which lasted for about 17 min. Because of the delay in starting the sodium pool fire, this experiment was not exactly comparable to Runs 303 and 304; in some respects, it was similar to the delayed mixing experiments (Runs 301, 302 and 306). The first aerosol mass sample was taken at 5 min into the sodium oxide generation period, and the mass ratio of sodium oxide to uranium oxide was 3.4 to 1. The second aerosol sample was taken 15.5 min into the sodium oxide generation period, and the mass ratio of sodium oxide to uranium oxide had increased to about 275 to 1. Early fallout of the sodium oxide aerosol removed a major fraction of the uranium oxide aerosol from the vessel atmosphere. The airborne uranium oxide content of the vessel atmosphere was reduced about 97% during the interval between the first and second aerosol sample. This observation was substantiated by the aerosol fallout rate measurements. At the termination of the experiment (48 hrs), the distribution of aerosol indicated that 84% of the sodium oxide and 80% of the uranium oxide had deposited on the floor of the vessel, and 16% of the sodium oxide and 20% of the uranium oxide had deposited on internal surfaces of the vessel.

Comments - Based upon the results from these six experiments, some general observation may be made. The airborne concentration of an agglomerated (aged) aerosol is reduced significantly when mixed with a newly-generated aerosol of source-size particles. For a given aerosol, either sodium oxide or uranium oxide, less material is made airborne when generated simultaneously with a second aerosol, than when generated alone. In both types of experiments on mixed-oxide aerosols, enhanced fallout is noted during, and shortly following, the generation or mixing periods. The aerodynamic behavior of a mixed-oxide aerosol seems to be similar even though the mass ratio of the two aerosols may differ at the time of mixing. In Fig. 6, all six of the experiments are compared. For this figure, time is measured from the start of mixing of the two aerosols so that the three delayed mixing experiments can be compared

with the three experiments where mixing was more or less simultaneous. The rate of disappearance of these mixed-oxide aerosols from the vessel atmosphere is similar even though the mass ratio and size particles at mixing vary significantly. All experimental evidence seems to indicate that the two aerosols co-agglomerate and the material remaining airborne acts in a manner similar to both sodium oxide or uranium oxide, but not entirely like either.

COMPARISON OF EXPERIMENTAL RESULTS WITH HAARM-3 CODE PREDICTIONS

The HAARM-3 aerosol code, being independently developed for NRC by Battelle-Columbus Laboratories, is utilized in the NSPP program for pretest predictions and posttest comparisons of the aerosol experiments. Comparisons between the code predictions and experimental results for sodium oxide aerosols and low-concentration uranium oxide aerosols have been generally favorable.[4,5]

The code predictions for recently-performed higher-concentration uranium oxide aerosol experiments (NSPP Runs 204-207) have also been favorable. For example, Fig. 7 illustrates the agreement for Run 207; the code prediction agrees with the experimental results within a factor of 2. Parameters used in the code input were: α = density correction factor = 1, χ = dynamic shape factor = 3, and γ = collision shape factor = 10.

Use of the HAARM-3 code to predict the behavior of a mixed-oxide aerosol is in an early stage of application at ORNL. Since the code was developed for a single-component aerosol, application in describing the behavior of a mixed-oxide aerosol requires appropriate averaging of the aerosol properties of the individual components for use as code input. As a first attempt, the mixed-oxide aerosols were treated as if they were single-component aerosol with a mass-averaged material density:

$$\bar{\rho} = (C_1\rho_1 + C_2\rho_2)/(C_1 + C_2)$$

where

$\bar{\rho}$ = assumed average material density for the two-component aerosol,

C_1 = initial mass concentration of U_3O_8 aerosol in the NSPP vessel $\mu g/cm^3$,

ρ_1 = U_3O_8 material density = 8.3 g/cm^3,

C_2 = initial mass concentration of Na_2O aerosol, $\mu g/cm^3$, and

ρ_2 = Na_2O material density = 2.27 g/cm^3.

The calculations were initiated at a time when the aerosol particle size distribution had been measured after the aerosol generation was terminated. A summary of the input data used in the calculations is presented in Table II.

Table II

A Summary of Input Data Used in HAARM-3 Calculations For
NSPP Runs 207 and 303-305

Run No.	207	303	304	305
Aerosol	U_3O_8	U_3O_8 + Na_2O	U_3O_8 + Na_2O	U_3O_8 + Na_2O
Initial time, min	25.8	28.5	25.3	36.0
Initial aerosol concentration,* 10^6 particles/cm^3	3670	7.77	25.0	5.23
Initial mass concentration, $\mu g/cm^3$	3.43	2.51	2.85	6.20
Initial aerosol mass median radius, μm	0.434	0.843	0.429	1.76
Initial aerosol geometric standard deviation based on radius	3.80	2.50	2.30	2.50
Mass-averaged aerosol material density, g/cm^3	8.30	5.63	7.81	2.27
Average vessel temperature, K	294	323	323	323
Temperature gradient at vessel wall, K/cm	negligible	negligible	negligible	negligible

*Estimated from the measurements of aerosol mass concentration and particle size distributions.

Figures 8 and 9 show the comparisons between the code predictions and the measured mixed-oxide aerosol concentrations for NSPP Runs 303 and 304 in which the uranium oxide mass was either nearly equal to or much higher than

that of the sodium oxide. Two calculations were performed for each run - one assuming that the behavior of the mixed-oxide aerosol would be like that of uranium oxide (with chain-agglomerate particles) and the other assuming that the behavior would be like that of sodium oxide (with nearly spherical particles). The code calculations based upon chain-agglomerate particle behavior compare more favorably with experimental measurements than those based upon spherical particle behavior.

Figure 10 contains the comparison for NSPP Run 305 in which the uranium oxide mass in the mixed-oxide aerosol was lower than that of the sodium oxide. The code calculation based upon chain-agglomerate particle behavior generally underpredicts, and the calculation based upon spherical particle behavior overpredicts the measured concentrations.

Based upon this initial application at ORNL of the HAARM-3 code for predicting the behavior of mixed-oxide aerosols, it appears that the mixed-oxide aerosol behaves more like an aerosol composed of chain-agglomerate particles, than spherical ones, when the uranium oxide mass is nearly equal to or higher than that of the sodium oxide. However, for a mixed-oxide aerosol in which the uranium oxide mass is much lower than that of sodium oxide, the behavior is neither like that of chain-agglomerate particles nor of spherical particles. Further investigation of this code application is underway.

ACKNOWLEDGEMENTS

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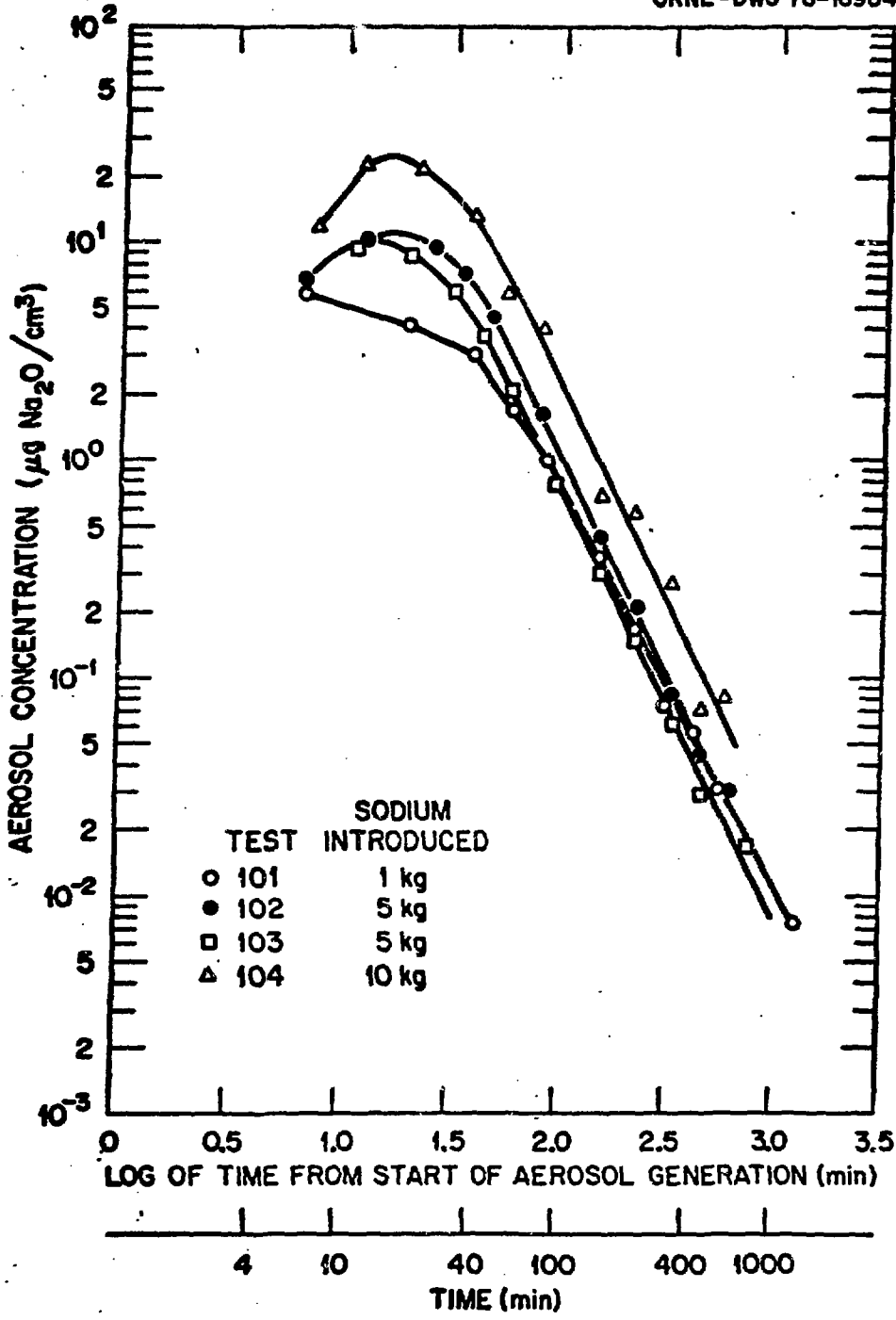
Fig. 1. Sodium Oxide Aerosol From Pool Fires
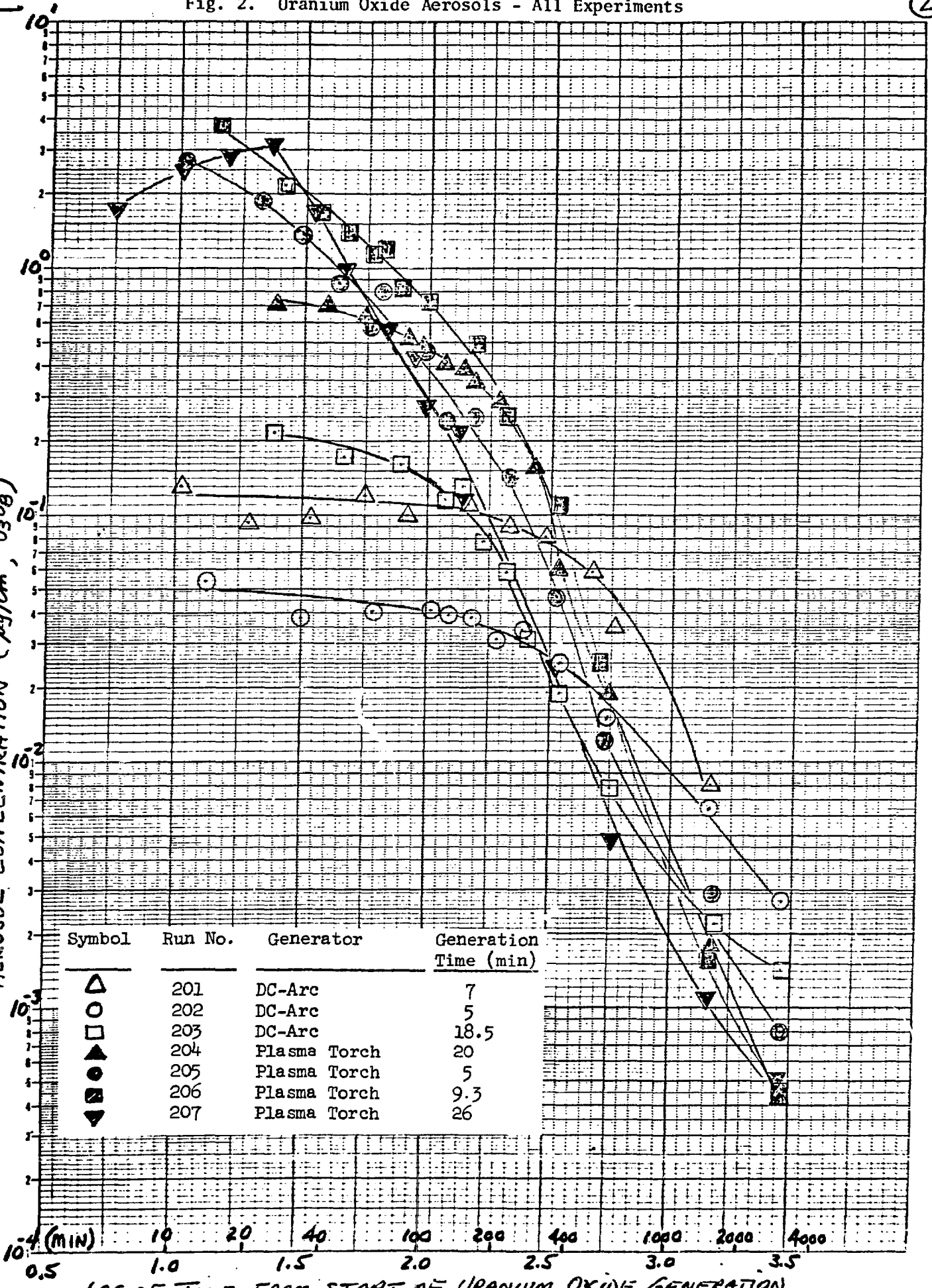
Fig. 2. Uranium Oxide Aerosols - All Experiments

Symbol	Run No.	Generator	Generation Time (min)
△	201	DC-Arc	7
○	202	DC-Arc	5
□	203	DC-Arc	18.5
▲	204	Plasma Torch	20
◑	205	Plasma Torch	5
▨	206	Plasma Torch	9.3
▼	207	Plasma Torch	26
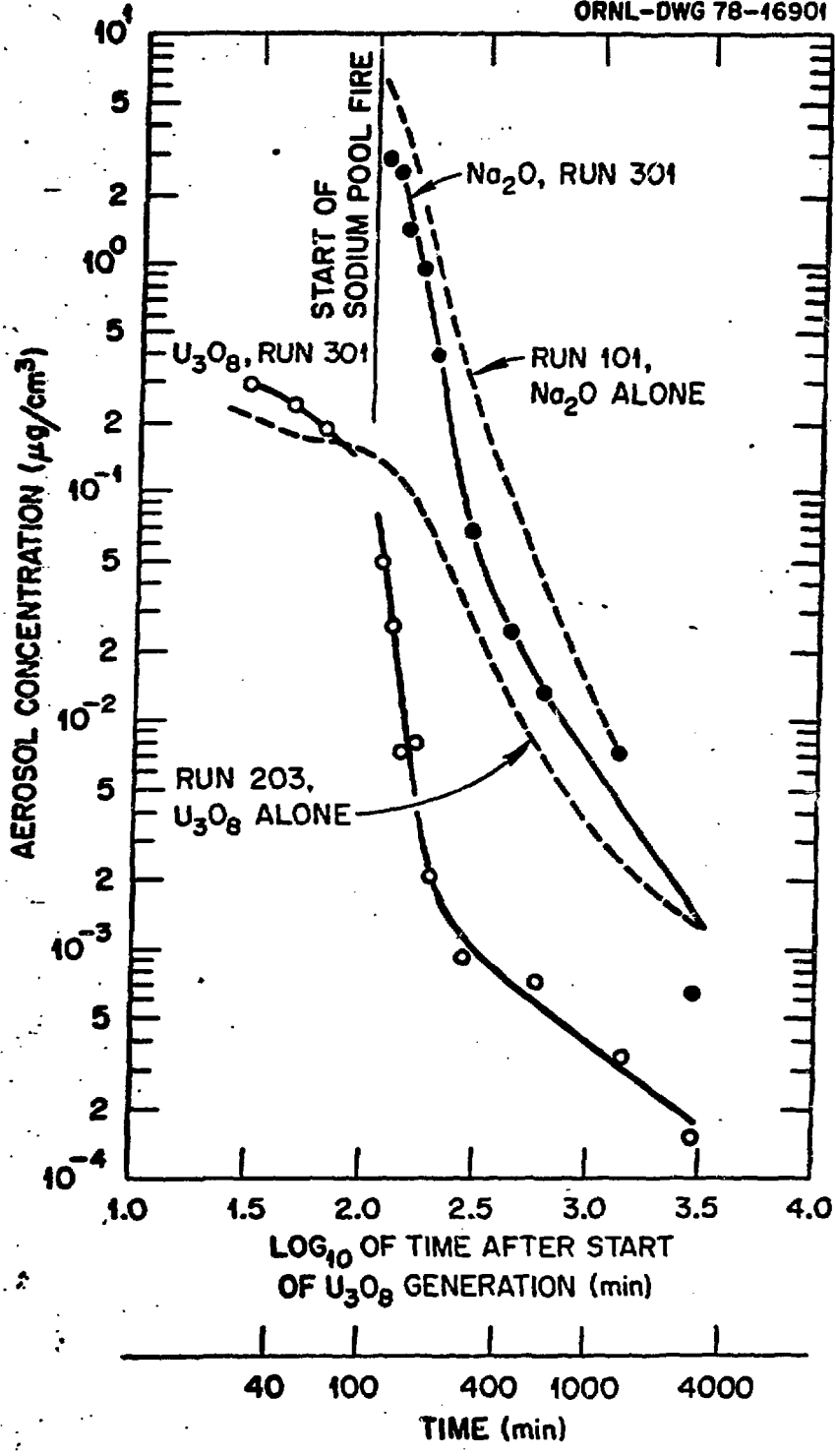
Fig. 3. Mixed Oxide Run 301

Fig. 4. Mixed Oxide Run 306

Label on vertical axis: AEROSOL CONCENTRATION ($\mu g/cm^3$, Na_2O or U_3O_8)

Graph annotations:
- Na_2O, RUN 306
- FOR COMPARISON: RUN 104 Na_2O AEROSOL ALONE
- U_3O_8, RUN 306
- FOR COMPARISON: RUN 206 U_3O_8 AEROSOL ALONE
- END OF Na_2O GEN. PERIOD
- U_3O_8 GEN. PERIOD

Horizontal axis: (min) ... FROM START OF SODIUM OXIDE GENERATION (MIN)

Fig. 5. Mixed Oxide Run 303

U_3O_8 COMPONENT

Na_2O COMPONENT

U_3O_8 GEN. PERIOD

Na_2O GEN. PERIOD
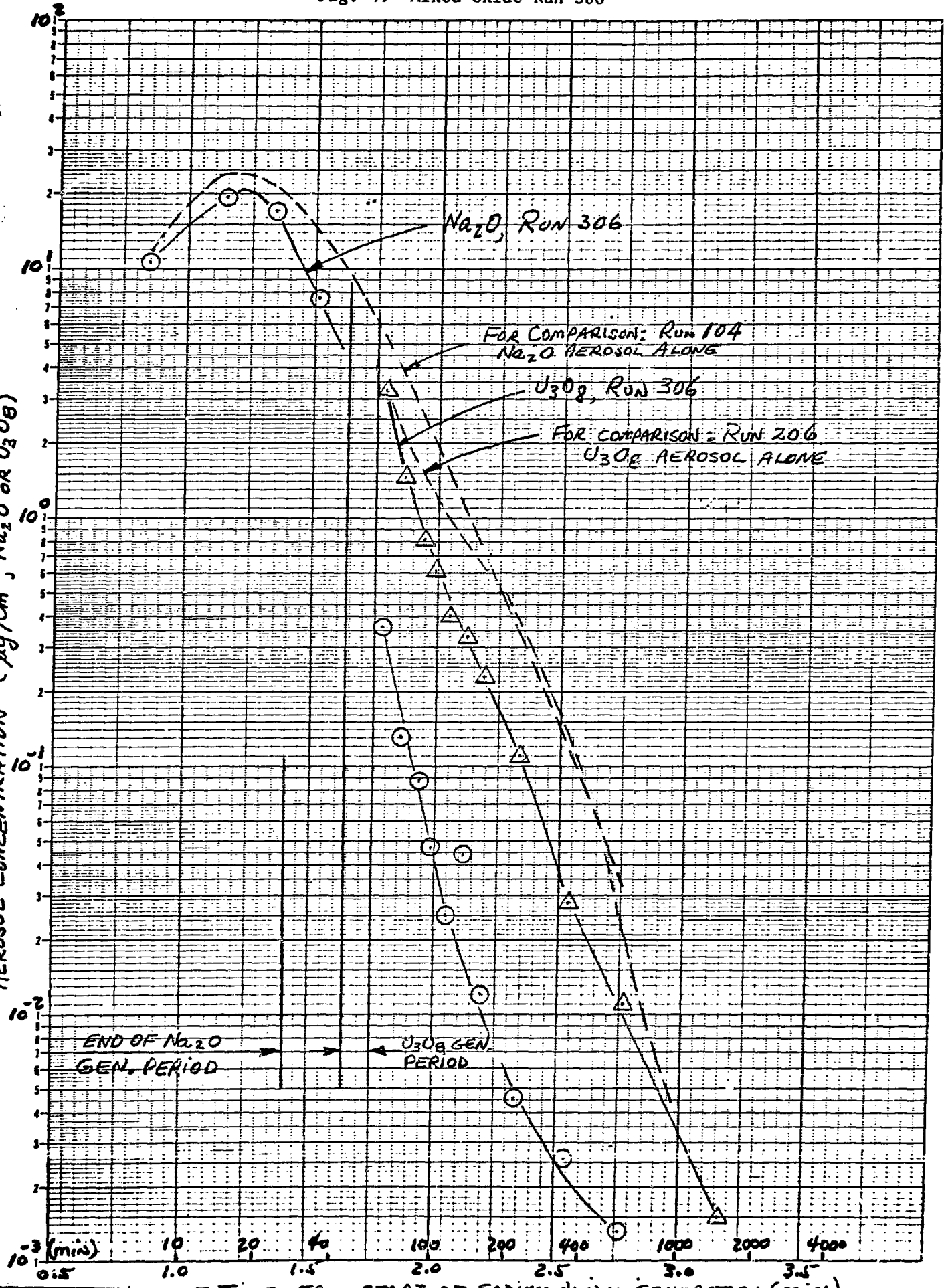
AEROSOL CONCENTRATION ($\mu g/cm^3$, Na_2O OR U_3O_8)
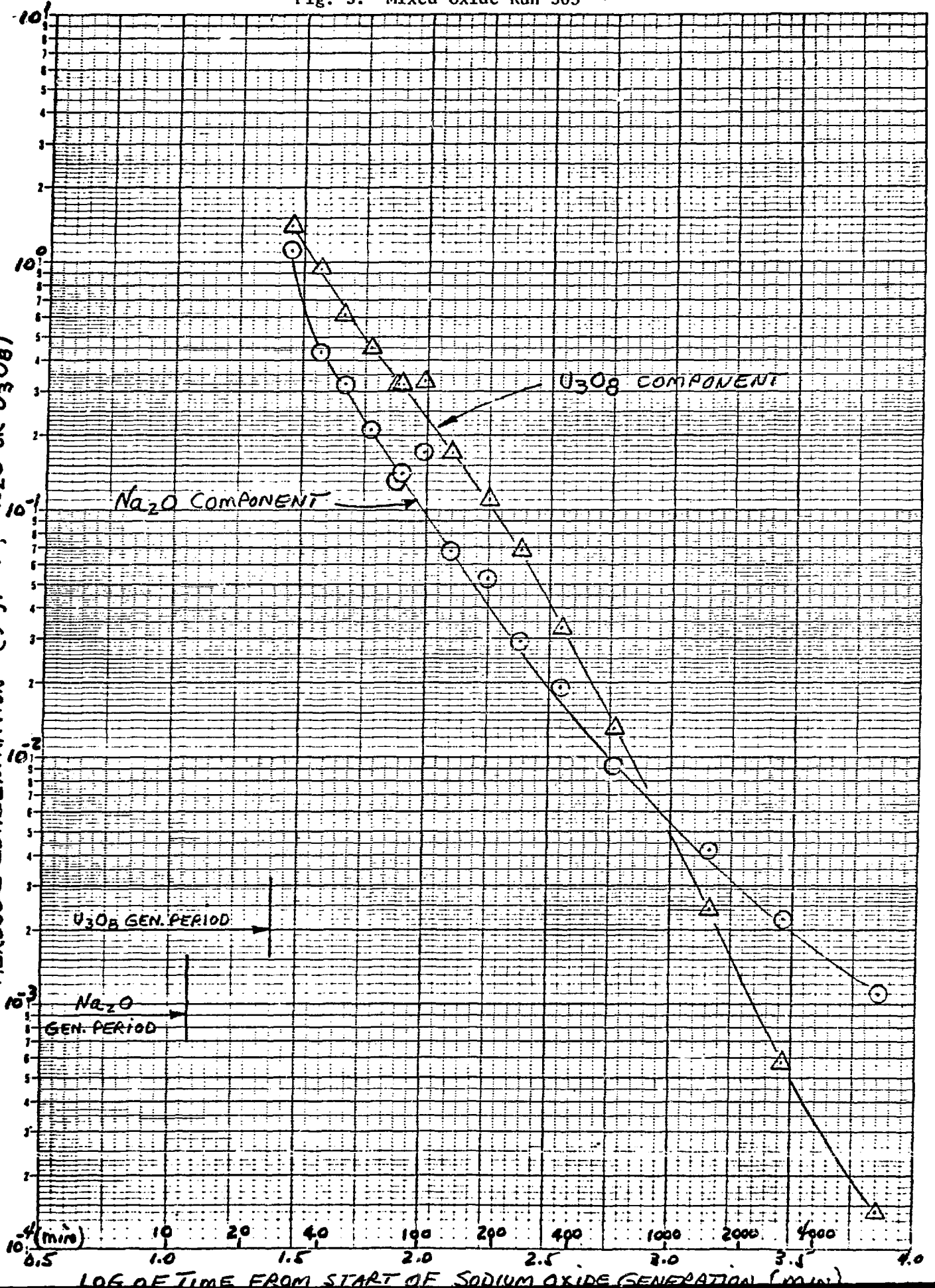
LOG OF TIME FROM START OF SODIUM OXIDE GENERATION (MIN)

Fig. 6. Mixed Oxide Runs 303, 304, 305

Symbol	Run No.	Aerosol Mass Ratio (Na_2O/U_3O_8)
◯	301	30/1
△	302	4/1
□	303	1/1
◇	304	1/10
⊘	305	3.5/1
▽	306	1/1

AEROSOL CONCENTRATION $(\mu g/cm^3, Na_2O + U_3O_8)$

Fig. 7. HAARM-3 vs. Test 207

⑦
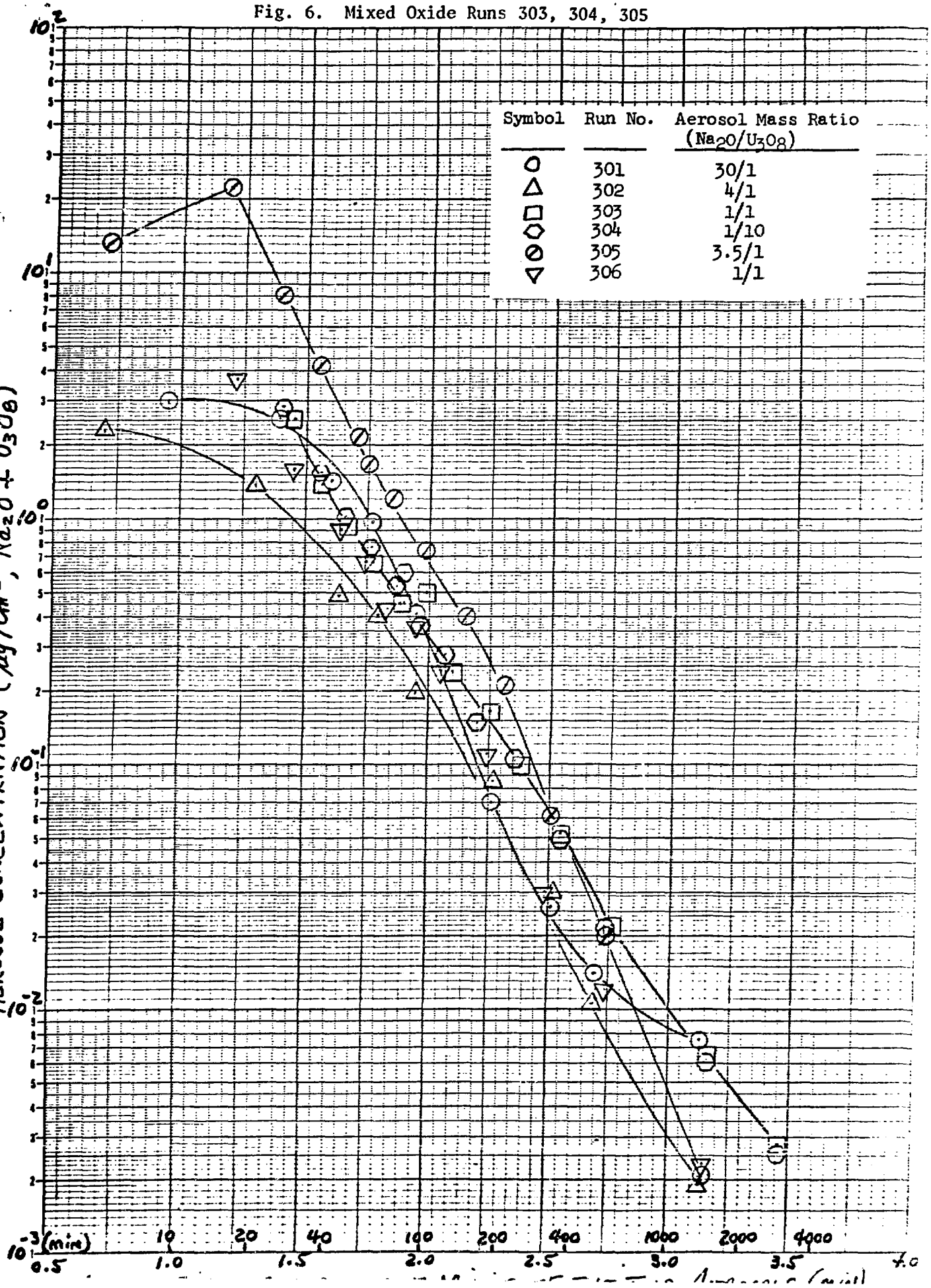
Fig. 7. HAARM-3 vs. Test 207
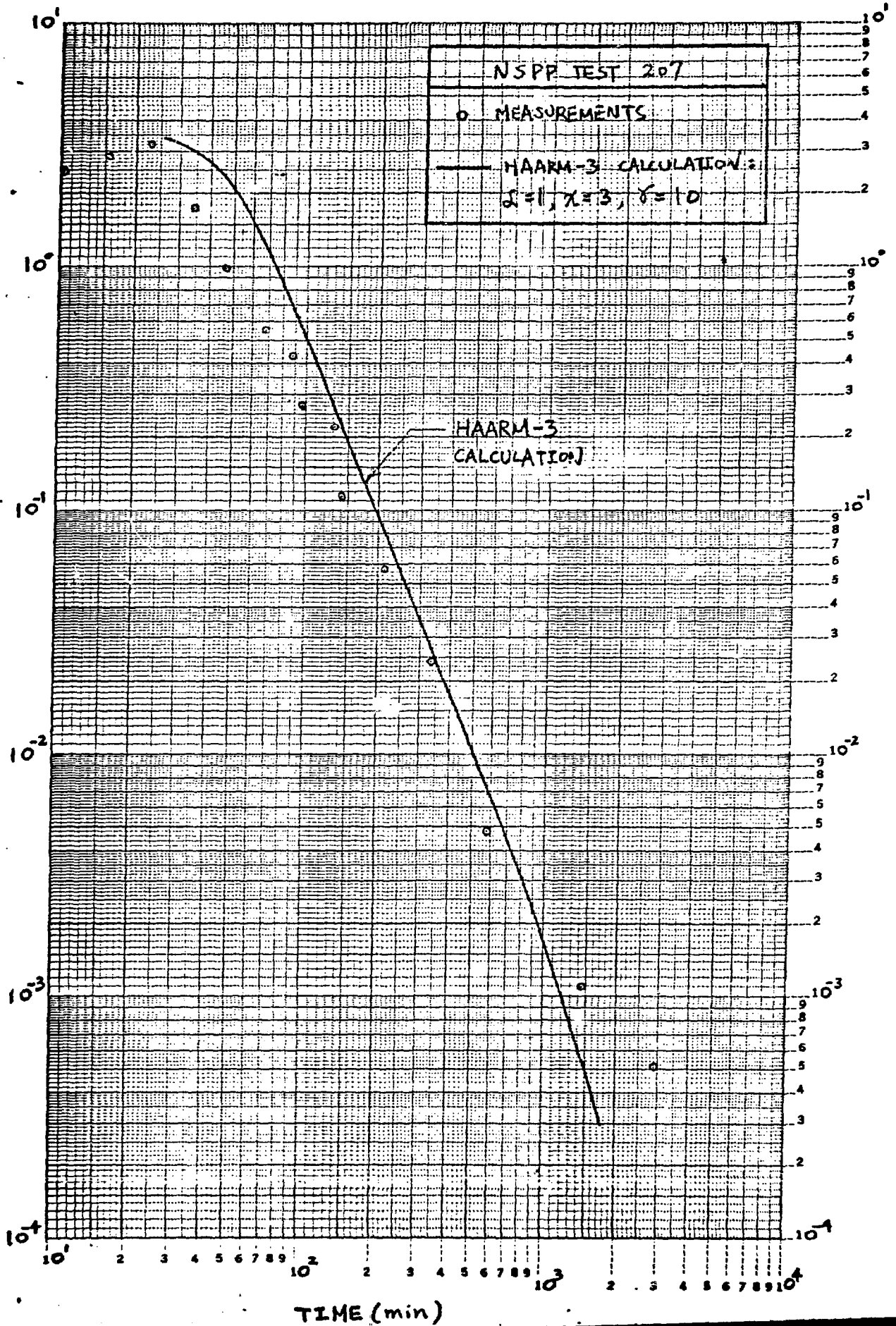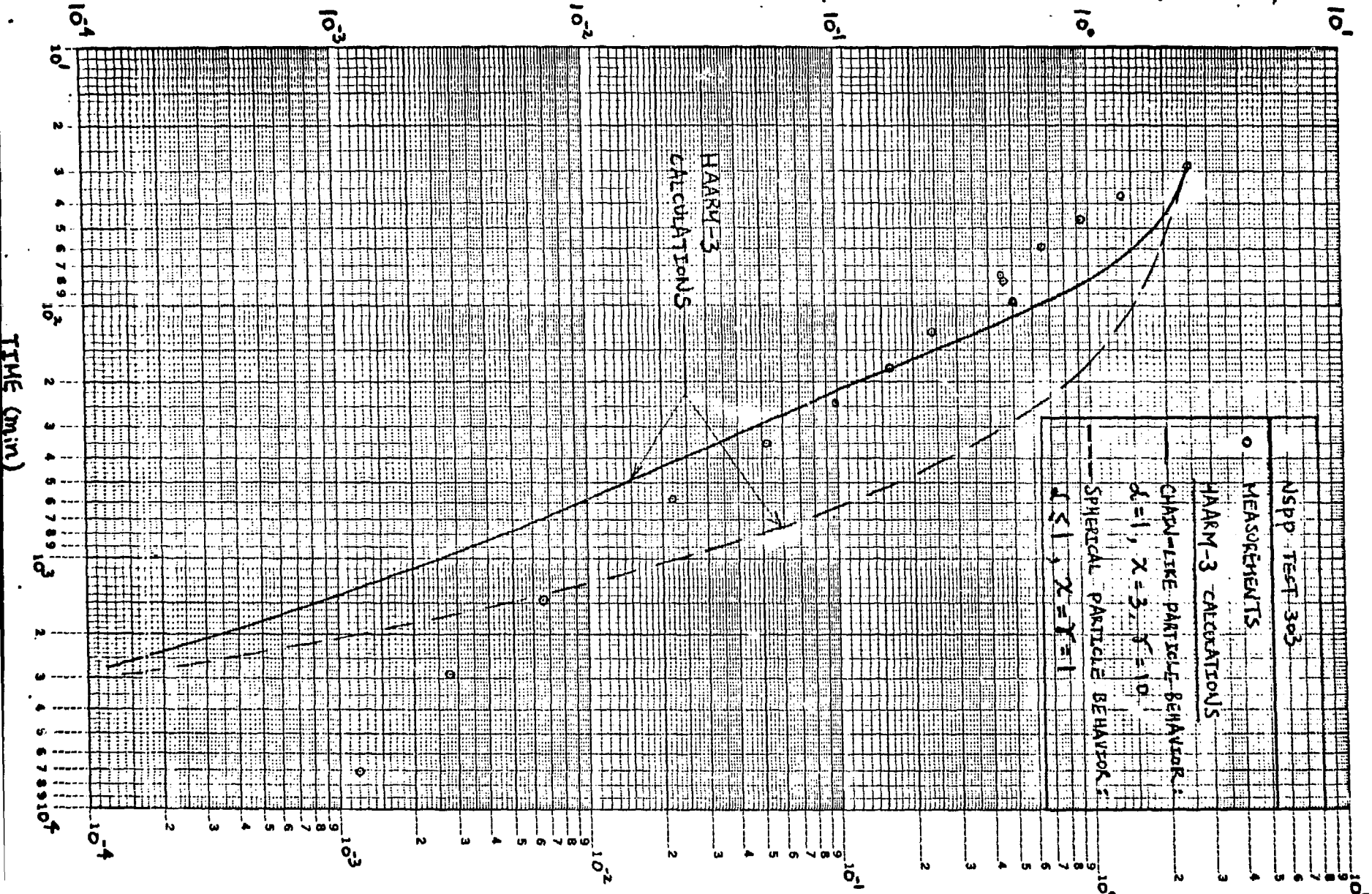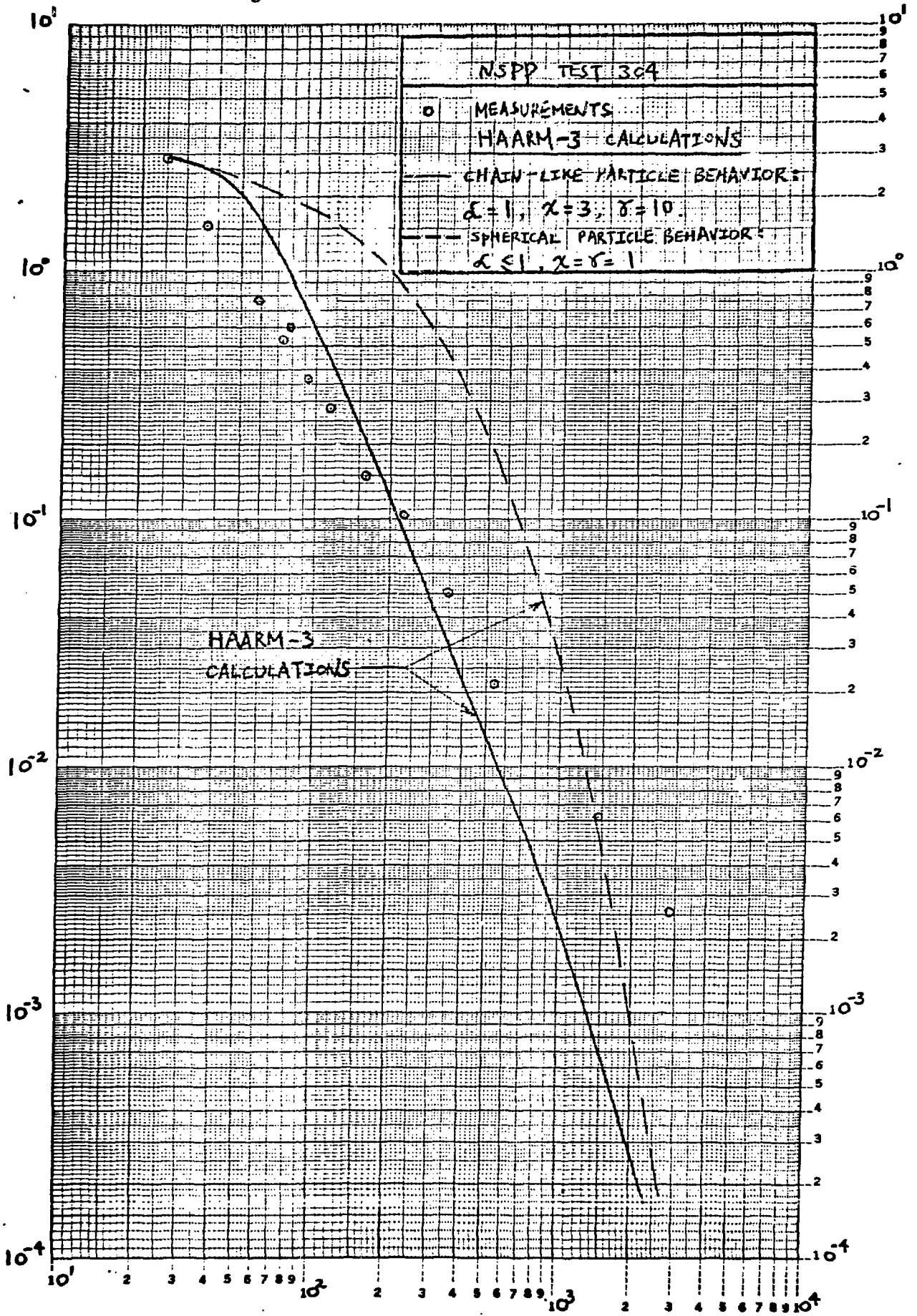
AEROSOL CONCENTRATION (Mg of U_3O_8 & Na_2O/cm^3).

TIME (min)

Fig. 8. HAARM-3 vs. Test 303

NSPP. TEST 303

o   MEASUREMENTS

HAARM-3 CALCULATIONS

CHAIN-LIKE PARTICLE BEHAVIOR:
$\alpha = 1$, $X = 3$, $\gamma = 10$

SPHERICAL PARTICLE BEHAVIOR:
$\alpha \leq 1$, $X = \gamma = 1$

HAARM-3
CALCULATIONS

Fig. 9: HAARM-3 vs. Test 304

Fig. 10. HAARM-3 vs. Test 305