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MASTER

THE BEHAVIOUR OF NUCLEAR AEROSOLS IN CLOSED SYSTEMS

Specialist Meeting in Karlsruhe on November 11 and 12, 1969

Compiled by
W. Schikarski

GESELLSCHAFT FUR KERNFORSCHUNG M.B.H.

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Juni 1970 KFK 1206

Institut für Angewandte Reaktorphysik
"Projekt Schneller Brüter"

THE BEHAVIOUR OF NUCLEAR AEROSOLS IN CLOSED SYSTEMS

Specialist meeting at the Nuclear Research Center
Karlsruhe, on November 11 and 12, 1969

by

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FOREWORD

On November 11 and 12, 1969 the "Specialist Meeting on the Behaviour of Nuclear Aerosols in Closed Systems" was held at the Nuclear Research Center Karlsruhe. The meeting was sponsored by the Institut für Angewandte Reaktorphysik of the center. About 20 participants from the USA, Japan, the Netherlands and Germany were present. It was the first international meeting of the special subject of nuclear aerosols which can be formed during accidents in nuclear installations. The aim of the meeting was the exchange of theoretical and experimental results, the discussion of the mechanism and processes involved in the behaviour of nuclear aerosols and the discussion of the various experimental and instrumental techniques used by different research groups. In particular, the application of the results on the problem of activity release during heavy accidents in fast sodium cooled reactors was discussed with some bearing on the problems of other nuclear reactor types.

Nuclear aerosols can be formed due to minor or major accidents in nuclear installations. The general behaviour is mainly determined by the parameters under which they are created, by the environmental conditions in which they are produced and by the volume, surface and other geometrical characteristics which govern the space in which they are suspended. The complex situation in a nuclear installation following a nuclear accident prohibits the reliable prediction of the behaviour of nuclear aerosols by theory. Theoretical models together with good simulating experiments are the tools to gain experience and knowledge of the behaviour of nuclear aerosols.

Since the meeting was a working one, most time was spent discussing the various problems. The most important topics, however, were introduced by short comprehensive papers presented by the different research groups. These papers are compiled in the following. The conclusions drawn from the discussions are listed at the end of this report.

W. Schikarski

Vorwort

Am 11. und 12. November 1969 fand in Karlsruhe das "Specialist Meeting on the Behaviour of Nuclear Aerosols in Closed Systems" statt. Die Tagung wurde vom Institut für Angewandte Reaktorphysik im Kernforschungszentrum Karlsruhe veranstaltet. Etwa 20 Teilnehmer aus den USA, Japan, Holland und Deutschland waren vertreten.

Die Tagung war die erste internationale Arbeitstagung für nukleare Aerosole und diente dem intensiven Gedanken- und Erfahrungsaustausch der die speziellen Fragen des Verhaltens nuklearer Aerosole bearbeitenden Fachleute. Unter dem Begriff "nukleare Aerosole" werden solche Schwebstoffe verstanden, die bei einem schweren Störfall eines Kernreaktors oder einer kerntechnischen Anlage auftreten können. Im einzelnen war es das Ziel der Tagung, die bei den verschiedenen Arbeitsgruppen in Japan, den Niederlanden, USA und Deutschland erzielten Meßergebnisse und die verwendeten Versuchs- und meßtechnischen Methoden zu vergleichen und zu diskutieren. Im Vordergrund stand die aktuelle Frage der Anwendung der bisherigen Ergebnisse auf das Aktivitätsfreisetzungs-Problem im Störfall natriumgekühlter Reaktoren, wobei die Übertragbarkeit der Ergebnisse auf andere Reaktortypen am Rande besprochen wurde.

Nukleare Aerosole können im Störfall von kerntechnischen Anlagen entstehen. Ihr Verhalten wird hauptsächlich von den Parametern, unter denen sie erzeugt werden, von den atmosphärischen Bedingungen, in die hinein sie erzeugt werden, und von den räumlich-geometrischen Verhältnissen, in denen sie luftgetragen bleiben, bestimmt. Da sich die komplexen Bedingungen in einer kerntechnischen Anlage nach einem Störfall der exakten Beschreibung weitgehend entziehen, müssen modelltheoretische Überlegungen herangezogen werden, um die Zusammenhänge zu klären. Durch gezielte Simulations-Experimente lassen sich bestimmte Einzelfragen beantworten und offene Probleme der Modelltheorie lösen.

Der Schwerpunkt der Tagung lag auf der Diskussion. Die wichtigsten Problemkreise wurden jeweils durch einleitende Übersichtsreferate angesprochen, die von den verschiedenen Forschungsgruppen gehalten wurden. Diese Vorträge sind im folgenden zusammengestellt. Die von den Tagungsteilnehmern erarbeiteten Empfehlungen sind am Ende des Berichtes aufgeführt.

W. Schikarski

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Paper Nr. 1

Introduction and Evaluation of Experimental Results on Mass Concentration Time Functions of Nuclear Aerosols

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Introduction

In 1965 we first began to study the problem of nuclear aerosols. The problem came up during a study on activity release in large fast nuclear reactors. In trying to predict activity release in a liquid metal fast breeder accident we first took the conservative approach by using the well-known assumptions of the MCA of a water-cooled power reactor. However, two major differences made the problem difficult

- In the worst accidents of fast reactors considered at that time a partial or complete vaporization of the core and fuel was predicted
- Because of the non-compatibility of water and sodium the water spray and water cooling systems used in water reactor containment systems could not be adopted as engineered safeguards in liquid metal fast breeders.

Therefore the accident models at that time did look extremely pessimistic for the possible activity release in a severe LMFBR accident, namely

- the maximum release of the radioactive inventory from the core was assumed
- essentially no activity release decreasing safeguards were applicable except the containment system itself and possibly an off-gas filter system.

In calculating the radiation burden to the public we soon arrived at the point where we required two fairly leaktight containment shells in series to reduce the accident doses to tolerable values. In the course of these considerations we disliked the assumption that essentially all the core material was vaporized and stayed airborne over a long period of time in the containment to contribute to the activity release because this assumption meant that hundreds of kilograms or even tons would stay airborne over a longer time period. To clarify the situation we made a parameter study on the relation of the containment system and of activity release by using several assumptions. Pessimistic assumptions of activity release, optimistic assumptions with very low primary release of the radioactive inventory and, what we considered, realistic assumptions which we called the "aerosol model of activity release" /‾1_7. The unique features of this model have been discussed elsewhere /‾2_7. Therefore I may recall here only a few important points of the aerosol model, namely

- the aerosol model asks how much radioactive material can stay airborne in the containment after a large accident in nuclear reactors and what is the time function of decrease.
- the aerosol model asks not about the primary release of fission products or fuel isotopes from the core. It is, therefore, independent on any events, sequences of events in the coarse of the large accidents
- since the aerosol model assumes that most of the core material will form recondensation aerosols there will be only two species of activities ready to be released to the environment: the aerosols and the fission gases. All other activities will very likely be adsorbed or attached by chemical reaction to the aerosols.

A further important aspect of the aerosol model is the fact that aerosol behaviour and aerosol parameters provide much better access to experimental investigation and justification.

Evaluation of Experimental Results

The most important parameter in the aerosol model is the aerosol mass concentration and their behaviour with time. We have collected all the published data on experiments with various aerosols in closed vessels. In particular, we have compiled the aerosol mass concentration time functions available and have tried to compare the experimental results obtained by the various research groups. It should be mentioned that the first curves of this type have been measured by Atomics International /‾3_7 and by Brookhaven National Laboratory /‾4_7.

The following research groups have made experiments on the time functions of aerosol mass concentrations

- Atomics International /‾3_7
- Brookhaven National Laboratory /‾4_7
- Reactor Centrum Nederland /‾5_7
- Kernforschungszentrum Karlsruhe /‾6_7

The experimental results of these groups are given in the figure. In order not to confuse the figure we have omitted several other curves measured by the research groups mentioned. All the curves, however, should be included in the discussion. The important data related to the experiments have been listed in the table.

The ten curves presented show a fairly disappointing situation. There is essentially no agreement between the experimental results of different laboratories if one disregards the fact that all measure a decrease in mass concentration vs time. Although all experiments show a decrease of mass concentration, the slope and shape of the time functions differ considerably. It is, therefore, necessary to evaluate the different experimental methods used. The following differences seem to be of influence

- type of aerosol
- aerosol measurement technique
- aerosol formation technique
- vessel volume and inner surface area
- vessel height

The table shows the quantitative data of these parameters for the experimental runs evaluated. This list of parameters can already explain the differences in the mass concentration time function. This list also indicates which factors could affect the behaviour of the aerosols.

It is difficult to compare such different experiments in a sense of evaluating quantitative results. However, qualitative conclusions can be drawn as the following:

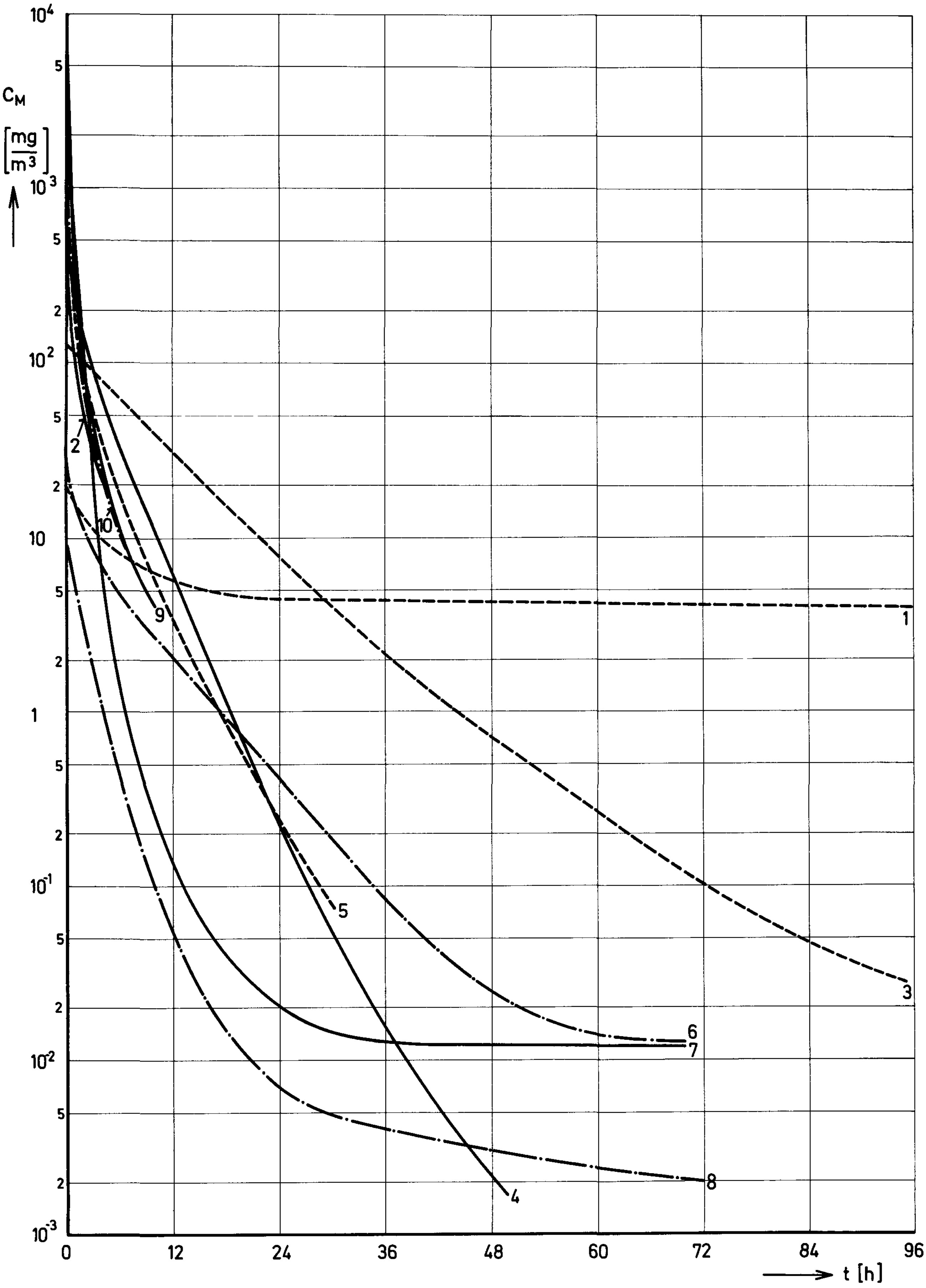
- airborne concentrations seem to remain longer suspended if the vessel height increases (curve 1 has the largest height, curve 3 has the second largest height). This, however, is not anymore valid for very small volumes (curve 7 and curve 6). It may be, that in those small volumes the height-effect (i.e. the sedimentation effect) is overshadowed by some other effect presumably diffusion.
- it seems to be generally true that for heigher initial concentrations the initial slope is much faster (curve 1 has the smallest initial concentration and has the slowest decay in the first hours of the mass concentration). This may indicate a strong influence of coagulation in the beginning.
- the question of whether there is a leveling off phase or not is split. The Brookhaven and the Karlsruhe results indicate that after a fast decay of roughly half a day a leveling off to a relatively stationary yet still decreasing mass concentration takes place. This effect was not observed by the Dutch group. Curve 3 indicates a relatively slow decrease with a nearly exponential behaviour throughout the time range and with no leveling off.

Since theoretical models appear to be not sufficiently developed we have not included calculations of the aerosol mass concentration time function. The complete understanding of the aerosol behaviour and the prediction of activity release necessitates both experimental and theoretical studies on nuclear aerosols.

Table

Comparison of experimental results on aerosol mass concentration vs time

Graph No.	Aerosol	Generation method	Energy kJ/g	Vessel atmosphere gas/pressure(at)	Initial concentration mg/m^3	Vessel Volume m^3	Vessel Hight m	Sampling or Measurement technique	Run No.	Reference
1	UO_2	Exploding wire	1	N_2 / 1 t	19,7	2,225	3,0	Filter Weighing	147	[6]
2	UO_2	"	1	N_2 / 1	1135	0,022	0,35	"	152	[6]
3	Gold	"	19	N_2	120	1,2	1,5	Activation analysis	ERGO 102	[5]
4	"	"	19	N_2	750	0,12	0,75	"	ERGO 8	[5]
5	Sodium	"	33	N_2	600	0,075	0,5	"	ERNA 14	[5]
6	PuO_2	resistance heating			50	0,75	1,0	α-counting		[4]
7	PuO_2	"			2000	0,017		"		[4]
8	7,5 % PuO_2 92,5 % Na	"				0,75		α-counting		[4]
9	UO_2 - 30 % Fe_2O_3 - 2,5 % WO_2 - 17 %	Arc generator			7000	1,0		impactor	test No. 3	[3]
10	UO_2 - 15 % Fe_2O_3 - 3,1 % Na_2O - 79,6 %	"			1200	1,0		"	test No. 4	[3]



Comparison of experimental results on aerosol mass concentration C_M VS time

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A SUMMARY OF RECENT PROGRESS IN AEROSOL RESEARCH AT BROOKHAVEN NATIONAL LABORATORY*

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Introduction

Safety considerations are of prime importance in making decisions concerning the design, siting, and operation of nuclear reactors. This is particularly true in the case of the liquid-metal-cooled fast breeder reactors (LMFBR's) which contain large inventories of radioactive fission products, sodium, and plutonium since the possible distribution of these materials into the environment presents a major potential hazard to the public. Due to the existing and proper USAEC philosophy of insuring public safety, unduly conservative licensing regulations will be imposed on reactor designers and the utilities in the absence of extensive technical information on the possible consequences of a power reactor accident.

A long range research program is under way at Brookhaven National Laboratory with the primary objective of developing a fundamental understanding of the chemical reactions, mechanisms of release, and transport behavior of the fission products and radioactive aerosols emanating from nuclear materials under conditions likely to be encountered in the event of an LMFBR accident. Recent progress pertaining to the aerosol physics phase of the program is discussed herein.

Knowledge of the nature of the radioactive aerosols likely to be produced in an LMFBR accident is important both in the design of engineered safety devices used to mitigate the consequences of a release and in making hazards analyses for deciding reactor containment and siting requirements. Although the general mechanisms involving agglomeration, settling and wall plating have been known for many years, the ability

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*This research was performed under the auspices of the U.S. Atomic Energy Commission.

to accurately predict the behavior of aerosols of nuclear materials under complex accident conditions is still in a stage of infancy. Experimental work is currently in progress to provide information on the nature of aerosols composed of PuO_2, UO_2, sodium, and combinations of each. In addition to the experimental work, a large theoretical research effort is under way to provide a basic understanding of aerosol behavior in closed systems. Computer codes are being developed for use in predicting the time dependence of the mass concentration, number concentration, and particle size distribution. These codes are used in data correlation and evaluation as well as for making scale-up calculations to realistic power-reactor accident conditions.

Experimental

Procedure - Most of the experiments carried out to date were made to investigate the size distribution and the time dependence of the mass concentration of aerosols vaporized into chambers with volumes of 0.8 m^3 and 0.02 m^3 having settling heights of 1.0 m and 0.3 m, respectively. The basic features of the apparatus have been described in detail elsewhere,(1) although several modifications have been made in generation and sampling techniques as described herein. All aerosols are now generated within the settling chamber to eliminate effects due to coagulation and removal which inevitably occur in transport lines. UO_2, PuO_2, and combination fuel oxide aerosols are produced using two different heating methods: (1) vaporization from a resistance heated TaC filament maintained at $\approx 2700^{\circ}C$, and (2) vaporization from a tungsten-lined TaC crucible inductively heated to temperatures ranging from 2700° to $3000^{\circ}C$. When required, sodium vaporization is accomplished using an auxiliary resistance heated nickel crucible maintained at $\approx 700^{\circ}C$.

Airborne particle sampling is done at three different heights in the larger chamber to ascertain whether good mixing is achieved. Thermal convection in actual accidents will undoubtedly lead to settling from a well-stirred environment and attaining this condition is one of the objectives of the current tests. Sampling of the aerosol is accomplished primarily by the use of hot-wire thermal precipitators. The aerosol is deposited on a carbon-coated copper electron microscope grid which is later examined with an electron microscope to determine particle size distributions. The grid is subsequently analyzed for mass concentration. Wall and floor deposition samples are obtained with a rotating-disk sampler which serves to expose coated electron microscope grids to the atmosphere of the enclosure for periodic intervals. Fixed plateout and floor sampling grids are also used to obtain the total mass-removal values for each process.

Results - The vapors of both PuO_2 and UO_2, as well as those of the mixed oxides, condense into small primary particles which readily coagulate into long branched chain aerosols. These branched chain aerosols are characteristic of those formed from metal oxide vapors.(2) Depending on specific experimental parameters to be discussed in more detail later, the aerosol chains form a wide range of sizes. These range from chains composed of only a few particles having apparent diameters around 0.01 micron to those composed of thousands of particles having apparent diameters of several microns. The individual particles comprising a chain are usually cubic but spherically shaped particles have also been observed. The formation of cubic particles is attributed to a vapor-solid phase transformation; nucleation to the liquid phase and then subsequent solidification probably results in the spherical particles sometimes observed. At the present time there is no evidence to suggest that coagulation is appreciably affected by the shape of the primary particle.

There is currently no "best" method of sizing aerosol chains. We use a Zeiss particle counter to size particles photographed from an electron microscope display of the deposition on the sampling grids. The reported radii are based on the equal-area circle-comparison technique. For short times after generation, the aerosols generally obey a log-normal size distribution so that specification of the total number density, the count-mean-radius (r_g) and the standard deviation, σ, uniquely determines the aerosol size distribution.

In our experiments, PuO_2 particles generated by vaporization at temperatures in the range 2500^o to 2900^oC were typically cubic in shape with individual particles in the size range 0.004 to 0.04 μ. Primary particle sizes were determined by sizing a chain of agglomerated particles. The distribution of these particles was essentially log-normal with a σ value ≈2.0 and a geometric mean particle size between 0.01 and 0.03 μ. Primary particle sizes for UO_2 aerosols were somewhat larger than for PuO_2 aerosols, although the σ values were approximately equal. This difference in primary particle size is most likely attributable to differences in the vapor pressure relationships of the two oxides (different PVT relationships during formation) rather than to different properties of the particles themselves. Visually, the two oxide-chain structures appeared to be nearly identical. Several experiments were made with 80% UO_2 - 20% PuO_2. These aerosols were also visually indistinguishable from those composed of either pure PuO_2 or pure UO_2, but again the sizes of the individual particles making up an agglomerate were generally somewhat larger than those typical of PuO_2 aerosols.

The initial r_g values for PuO_2 aerosols formed at nominal (based on weight of material vaporized) mass concentrations of 0.06 to 3.1 g/m^3 were generally within the range 0.05 to 2.0 μm. Corresponding σ values for the distributions usually averaged about 1.9 but values ranging from 1.5 to 2.1 have been observed. Similar values of r_g and σ have been measured for UO_2 aerosols generated with nominal mass concentrations as high as 9.0 g/m^3.

The production of combined oxide and sodium aerosols usually resulted in the formation of the characteristic oxide branched-chain agglomerate with the relatively large sodium spheres ($0.1 \geq r_g \geq 0.3$ μm) incorporated directly into the chain. The relative quantity of each primary particle, of course, depends on the sodium to oxide ratio of the individual experiment.

All of the experimental results indicated that the airborne mass concentration of the fuel oxide aerosols dropped rapidly during the first hour of settling per meter of settling-vessel height. Subsequently, the rate of aerosol settling decreased continually until the airborne concentration reached $\approx 10^{-6}$ g/m^3, a level which was sometimes observed to persist for several days. This undoubtedly results from the fact that some relatively small particles remain airborne following the rather rapid initial coagulation and settling regimes. These smaller particles settle much less rapidly and only slowly undergo further agglomeration due to their dilute concentration and therefore rather infrequent occurrence of collisions. In nearly all experiments the r_g value of the aerosol distribution was found to increase initially, approach a maximum, and then gradually decrease.

Only a few experiments were performed to study the effect of sodium on the behavior of the PuO_2 airborne mass concentration. Based on preliminary results, the presence of sodium was found to have some effect. For sodium to PuO_2 ratios of 1, the long-time behavior of the aerosol was very much like that observed without sodium. However, with large sodium to PuO_2 ratios, the aerosol was observed to remain airborne for a longer period of time. This observation is not unexpected. Since the density of sodium is quite low compared to PuO_2, the composite agglomerates, although larger in size, have a smaller density than the agglomerates of pure PuO_2. This should lead to a somewhat smaller value of the Stokes settling velocity and a more persistent aerosol.

Theoretical Considerations

It is a well known fact that high number density aerosols rapidly coagulate, and furthermore, experimental evidence indicates that coagulation results from nearly every collision between two particles.(2) As the particles coagulate, their sizes increase until the random thermal (Brownian) motion gives way to preferential settling due to gravitational forces, and the particles settle-out. In addition, particle transport by convection currents and diffusion results in their transfer to and plateout on other surfaces of the enclosure.

A computer code has been developed at BNL for solving the partial differential-integro equation expressing the important mechanisms influencing the aerosol behavior. The code, TRAP, (Transient Removal of Aerosol Particles) includes terms to account for coagulation by Brownian motion, gravitational, and turbulent agglomeration, and decrease in airborne concentration by wall plating, leakage, and Stokes settling.

A source term is also included to account for aerosol generation over a finite period of time. The details of the formulation of the basic kinetic relationships, as well as the code development and the method of solution, are given elsewhere.(3)

Application of existing computer programs has been severely limited since the initial particle size distribution is required for their use in analyzing practical problems. Although numerous measurements have been made for specific test conditions, no useful method for predicting short-time size distributions has heretofore been available. Recently, we developed a method for calculating the instantaneous-source size spectrum of an aerosol in terms of its initial mass concentration.(3,4) This was based on the self-preserving distribution developed by Swift and Friedlander.(5)

The development begins with the size distribution function where

$$n(v) = \frac{N_t^2}{\phi} \psi(\eta) \tag{1}$$

where

$$\eta = \frac{vN_t}{\phi} \tag{2}$$

$$\phi = \int_0^\infty n(v)\,dv \quad . \tag{4}$$

It is assumed that although the number concentration and hence the mean particle size varies during coagulation, the distribution shape, $\psi(\eta)$, is time independent. In Eqs. (1) to (4), $n(v)$ is the number of particles per unit volume in the size range dv, t is time, and N_t and ϕ are total number and total volume of particles per unit volume, respectively. Combining these equations with the log-normal distribution, which has been found to be an excellent approximation to the short-time aerosol size distribution, leads to

$$\phi = \frac{4\pi}{3} N_t \, r_g^3 \exp\left(4.5 \ln^2 \sigma\right) \tag{5}$$

$$\eta = \left(\frac{r}{r_g}\right)^3 \frac{1}{\exp(4.5\ \ell n^2\ \sigma)} \tag{6}$$

$$\psi = \frac{1}{3\eta}\ \frac{1}{\sqrt{2\pi}\ \ell n\ \sigma}\ \exp\left(- \left\{ \frac{\ell n[(\eta)^{1/3}\ \exp\ 1.5\ \ell n^2\ \sigma]}{\sqrt{2}\ \ell n\ \sigma} \right\}^2 \right) \tag{7}$$

For an arbitrary log-normal distribution of specified σ, both η and $\psi(\eta)$ are functions only of r/r_g. Therefore if $n(v)$ has a self-preserving log-normal shape, σ must be a specific constant, independent of time.

Aerosol generation by condensation of the super-saturated vapor is the likely mechanism of aerosol formation in the event of a fast reactor accident. An aerosol produced by this mechanism would be highly dispersed with a large number density, and would probably have idividual particles with radii ranging between 20 and 100 Å.(6) Aerosols with $N_o \geq 10^9$ particles/cc coagulate rapidly into larger particle chains before removal processes become significant. Using a CDC 6600 digital computer, we solved the basic aerosol equation to determine whether a self-preserving particle spectrum results after significant coagulation. Calculations were performed for PuO_2 and UO_2 aerosols having initial log-normal distributions with r_g ranging from 20 to 100 Å and σ varying from 1.2 to 2.5. Initial mass concentrations and number densities ranged from 10^{-2} to 100 μg/cc and 10^9 to 10^{14} particles/cc, respectively. Prior to times when particle removal becomes significant, the number density distribution remained very close to log-normal with σ rapidly leveling off to values between 1.34 and 1.40 for all cases. Thus for the purpose of calculations, the initial size distribution of high mass concentration aerosols can be approximated by a log-normal distribution with σ of 1.37. This leads directly to a specific r_g in terms of mass concentration:

$$C_o = 6.54 \times 10^8\ \rho r_g^3 \quad . \tag{8}$$

Eq. (8) was used to determine the initial particle size distribution for a series of parametric studies. The effect of initial concentration on the time dependence of the fractional mass concentration remaining airborne is shown in Figure 1 for initial concentration values ranging from 0.01 to 100.0 g/m^3, a settling chamber height of 100 cm, and a particle density of 6.0 g/cc. The mass concentrations are shown in the figure together with the corresponding values of the initial geometric mean radii, expressed in microns. As would be expected, the higher the mass concentration and corresponding initial radii, the more rapid the decrease in the fraction of the mass remaining airborne. Note that this is for the case of an instantaneous source.

The present results show that with simultaneous coagulation and settling, the geometric mean particle radius increases with time, approaches a maximum, and then decreases to a nearly constant value. This peaking in radius has also been observed experimentally.

Other computations were made to study the effect of vessel height on the aerosol settling. Figure 2 shows the fraction of aerosol mass remaining airborne as a function of time for a case with an initial concentration of 10.0 g/m^3. Settling results are plotted for chambers with heights of 10, 30, 100, and 1000 cm. Although increasing the vessel height results in an increase in time for a given fraction of the mass concentration to settle, the effect of height is less in the case of simultaneous coagulation and settling than for the case of Stokes settling only.

Comparison of Experiment with Theory - A comparison of theory with some of our experimental results is shown in Figures 3 and 4 where the reduction in aerosol concentration with time is plotted for several PuO_2 and UO_2 runs. The ordinate of the figures is the fraction of aerosol remaining in the enclosure and it was obtained by normalizing the experimental and theoretical concentrations to the initial value. Calculations for PuO_2 and UO_2 aerosols assumed an initial σ of 1.9 and an aerosol density of 6.0 g/cc. This was found to be a good value for the effective density and adequately accounted for a lowering in the Stokes settling-velocity due to: (1) void spaces in the aerosol chains and (2) increased drag forces for the settling of chains compared to spheres. The mechanisms of Brownian, sedimentation, and turbulent coagulation, and settling and wall plating were included in the calculations.

Although the aerosol vaporization was generally quite rapid, the experimental conditions did not fulfill the requirements for an instantaneous source, and therefore, Eq. (8) was not used to evaluate the initial r_g. Instead, experimentally measured r_g values obtained 3 to 5 minutes after cessation of vaporization were used. A σ of 1.9 represents an average of the values obtained in all of our experiments and this was used in making the computations.

Referring to Figure 3, it can be seen that the calculated results are in very good agreement with the experimentally obtained concentrations. Since the initial experimental parameters for runs PuO_2-a and PuO_2-b are very similar, as are the experimentally obtained variations of concentration with time, this leads to the conclusion that the aerosols are behaving in a reproducible manner.

A comparison of theory and experiment for the UO_2 runs shows that the agreement is not quite as good as for PuO_2 experiments. UO_2-a and UO_2-b had similar initial conditions but behaved in a somewhat different manner experimentally. This may reflect the inability to analyze small concentrations of UO_2 as accurately as similar concentrations of PuO_2; the latter are analyzed by α-counting. It can be noted that the transient concentration curves predicted for these two cases are almost identical; however, the UO_2-b experimental points generally fall above whereas the UO_2-a experimental points generally fall below the predicted curves. Run UO_2-c had the highest concentration and the analysis of the concentrations for this run is probably better than that of UO_2-a and UO_2-b. For early times, the theoretical curve lies slightly below the experimental data, whereas at later times, the reverse is true.

Conclusions

Experimental results are available on the nature of PuO_2 and UO_2 aerosols up to intermediate nominal mass concentrations of several grams per cubic meter. The BNL code TRAP is seen to be quite adequate for describing the general behavior of PuO_2 and UO_2 aerosols at the concentration levels of the experiments. Because of the difficulty in carrying out aerosol experiments with these materials, the agreement between the calculated results and the experimental data is considered to be quite good.

Preliminary results on mixed fuel oxide-sodium aerosols indicate that the formation of low-density mixed agglomerates leads to a slower settling-rate and a somewhat prolonged time for which a given concentration of particles remains airborne.

The dominant aerosol processes are coagulation and settling; wall plating is of somewhat less importance depending on the surface to volume ratio of the vessel. Consequently, wall plating by diffusion should probably be negligible in an actual reactor, but other processes such as inertial impaction and thermophoresis may be important. The geometrical factor of major importance is that of the height of the settling vessel. However, for cases of high initial mass concentration, the effect of vessel height is less than expected considering Stokes settling alone.

Based on the approximation that coagulation results in the formation of spheres, the initial agglomerate size can be computed from considerations of the self-preserving distribution due to Friedlander. For the case where the initial size distributions are log-normal and the source is instantaneous, the particle size is inversely proportional to the cube-root of the mass concentration.

The experimental results, confirmed by theoretical calculations, indicate that the mass concentration of an aerosol decreases rapidly for the first hour per meter of settling-vessel height. Thereafter, the rate of change of the aerosol mass concentration continually decreases. A very low mass concentration aerosol has been observed to persist for several days. Although it is premature to attempt an exact quantitative comparison, the results obtained by BNL researchers and those at G.f.K. are in good qualitative agreement.

Acknowledgments

The author gratefully acknowledges the contributions of various members of the group to this research. The individuals include: P. Bezler, C. Brewster, F. Horn, G. Lindauer, and D. Wales. The author also thanks I. Tang for helpful discussions during the course of this work.

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Figure 1 -VARIATION OF CONCENTRATION WITH TIME FOR INSTANTANEOUS SOURCE MODEL

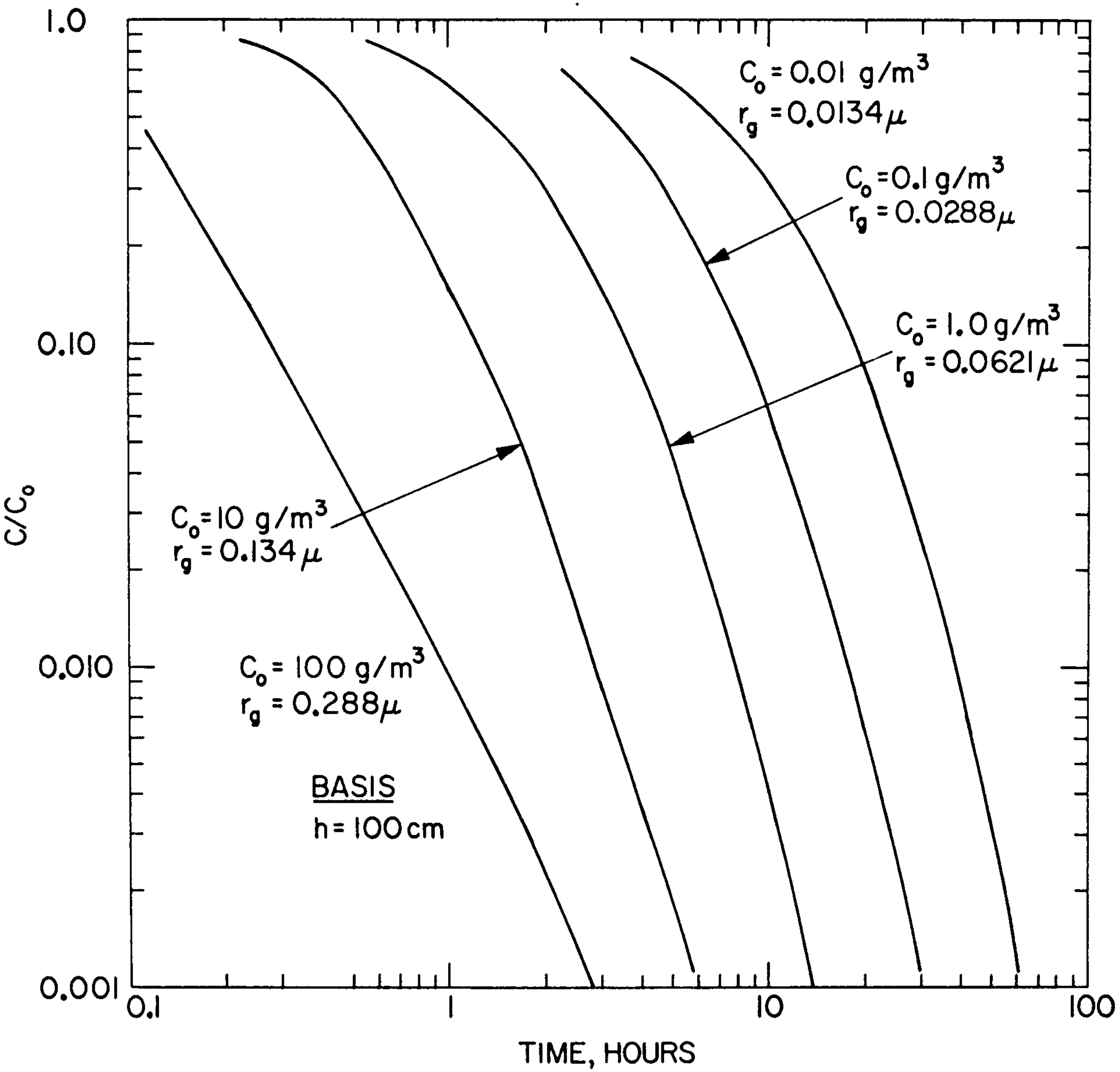


Figure 2.- EFFECT OF VESSEL HEIGHT ON AEROSOL CONCENTRATION FOR INSTANTANEOUS SOURCE MODEL

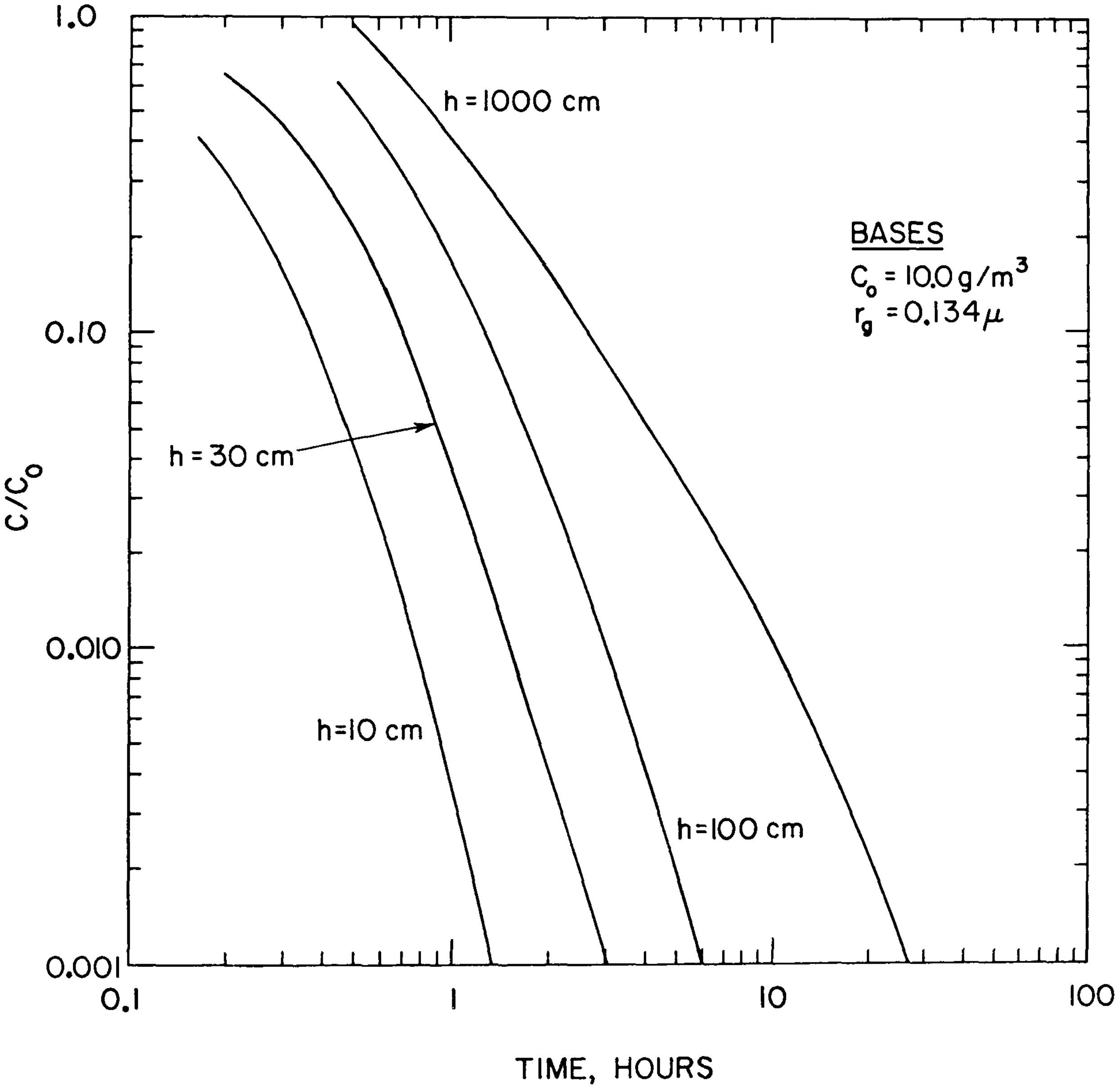


Figure 3 - VARIATION OF CONCENTRATION WITH TIME FOR PuO_2 AEROSOLS

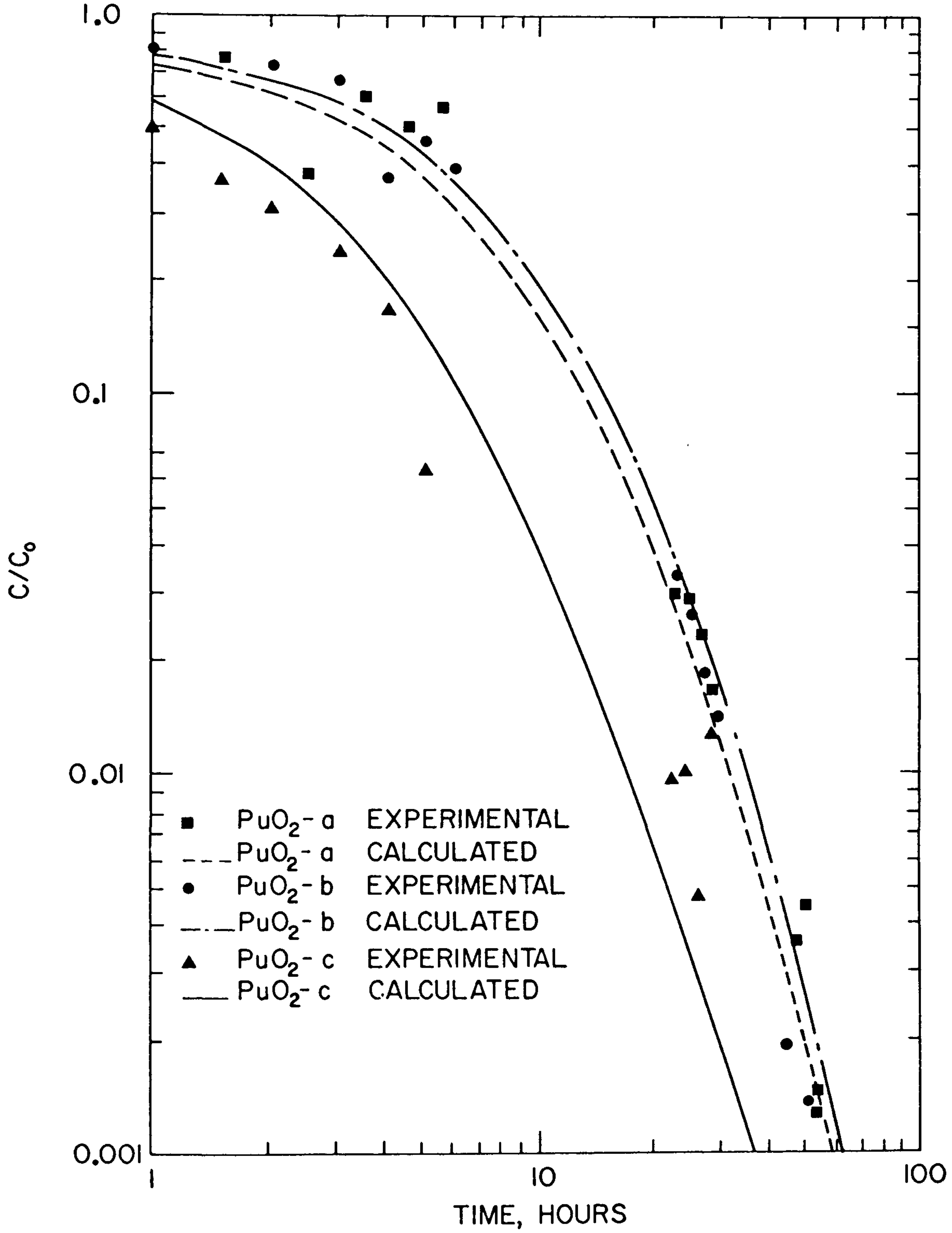
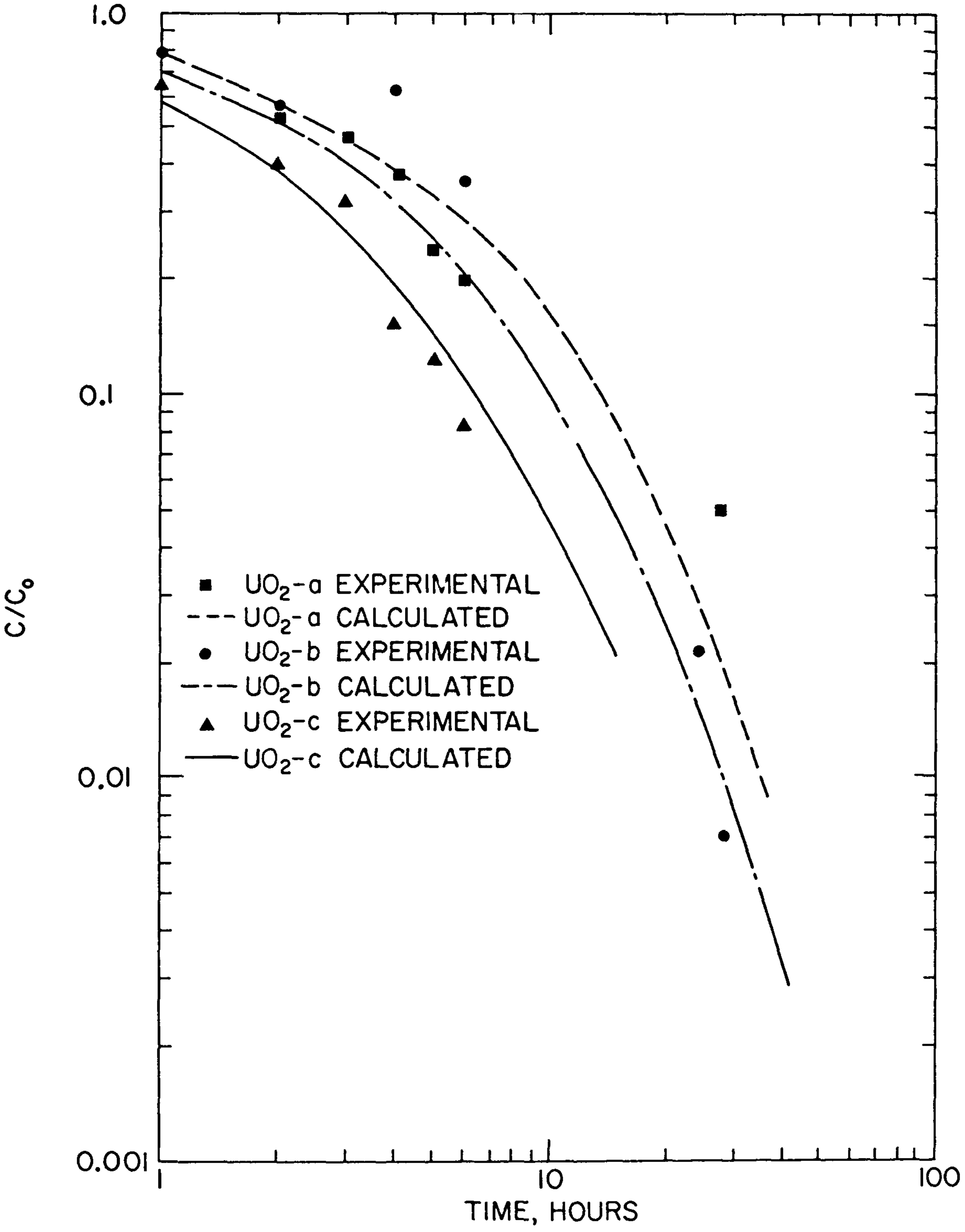


Figure 4 - VARIATION OF CONCENTRATION WITH TIME FOR UO_2 AEROSOLS



SOME ASPECTS OF NUCLEATION AND GROWTH*

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Discussion Paper Presented at the Specialist Meeting on the Behavior of Nuclear Aerosols in Closed Systems at Karlsruhe, Germany, November 11-12, 1969

Introduction

Some aspects of the processes of nucleation and growth are considered in this paper with the objective of suggesting some environmental parameters which may significantly influence particle sizes and hence airborne lifetimes. Experiments to study the effects of these parameters are schematically described.

The Size and Concentration of Primary Particles

Figure 1 shows some estimations of the sizes of UO_2 nuclei at the time of their birth, based on equilibrium concepts. The plot is of supersaturation (i.e., the ratio of the partial pressure of UO_2 to its equilibrium particle pressure), vs particle radius. The curve labeled "uncharged nuclei" is a plot of values from the well-known Kelvin equation.(1) The Kelvin equation expresses a metastable equilibrium. If nuclei are slightly larger than indicated by the Kelvin equation, then they grow spontaneously; if they are smaller than indicated by the equation, then they evaporate spontaneously. An estimated value of surface tension for UO_2 at its melting point was employed,namely 350 poise. Note that the critical particle radius values, given by the Kelvin equation, are very small. They are only several Angstroms.

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If the nucleus is charged, then the molecules in the nucleus tend to separate from each other. This electrical force operates against the surface tension and the apparent surface tension is decreased. A small nucleus is not likely to contain more than one charge and a single charge cannot be uniformly distributed on the surface. Therefore an effective charge distribution is considered which results from polarization. Consequently, the effect depends on the dielectric constant of the nucleus substance (which was estimated to be 10 for UO_2). A formula which relates supersaturation and radius of singly charged nuclei has been described in the book by Green and Lane(2) and appears in Fig. 1 under the notation "singly charged". This formula, like the Kelvin equation, describes an unstable equilibrium. Note that in this case there is a critical value of supersaturation, i.e., the maximum in the curve, above which all sizes of nuclei grow spontaneously. So if UO_2 nuclei are singly charged they will grow spontaneously at supersaturations above about 4, regardless of size.

Figure 2 introduces the question of the probability that a nucleus might be singly charged. The values of this curve were taken from the literature.(3) The graph shows the fraction of nuclei which are singly charged vs nuclei radius. These values are for an aerosol in equilibrium with an ion atmosphere containing the same concentration of positive and negative ions. It can be seen that the probability of Angstrom size particles being charged is very low.

There are two other points which may affect particle charging that I have no quantitative information about. One is that in a very high ion concentration, such as produced by a high radiation field, normally second order effects such as ion-dipole interactions may become important.

The other point, which seems very likely to be important, is that the ion atmosphere in the radiation field near a radiation source is not exactly bipolar because the beta particles have a longer range than the positive nuclei. In other words, there is some charge separation and the positive ion concentration may be higher than the negative ion concentration near the radiation source. It is suggested therefore that for nucleation near a vaporizing, highly radioactive source (that Fig. 2 will not apply), that many of the very small nuclei will be charged.

Figure 3 shows some estimates based on the kinetics of nucleation. This is a plot of the rate of generation of nuclei (no/cm^3-sec) vs supersaturation ratio. The equation shown on the figure was used to calculate the curves and it has been described.(4) The conceptual basis of the equation is that groups of molecules occur in the concentrated vapor as a result of statistical fluctuations. So J, the nuclei generation rate, is the rate of formation of groups of molecules which exceed the critical size given by the Kelvin equation (and henceforth grow spontaneously). In other words, we have here, a kinetic estimate of the rate of nuclei formation in the absence of electrical charge effects.

Figure 3 contains three estimates of nuclei generation rate for UO_2 near the melting point: one estimate assumes the surface tension is 200 dynes/cm, one assumes 350 and a third 500 dynes/cm. There are two estimates for Na_2O at 1550°K, one assumes the surface tension to be 200, and one assumes 300 dynes/cm. Values of J (nuclei generation rate) for water are given for comparison. Note that sodium apparently requires a very high supersaturation to self-nucleate, i.e., a few hundred compared to 1.5-6 for the other materials.

If there is an effect due to particle charges on nucleation rate such as previously suggested with regard to the equilibrium estimations, then it would be in the direction of increasing the nucleation rate, i.e., increasing the number of primary particles formed and decreasing their size.

It has been suggested in the foregoing development that primary particle size may depend on: (1) the supersaturation ratio in the nucleation zone, (2) on the surface tension of the substance nucleating, (3) on the temperature and (4) on radiation (i.e., on the ion atmosphere caused by radiation).

Let us now consider supersaturation further to see what environmental parameters determine it. A supersaturated vapor may be formed by cooling a saturated vapor or by chemical reaction. These two cases will be considered separately.

Figure 4 shows a model of the situation near a vaporizing surface where supersaturation develops by cooling a saturated vapor. At the top of the figure is a diagram showing a hot surface being swept by a stream of cool gas. There is a stagnant film of gas between the hot surface and the bulk of the gas stream. The plot is of several parameters vs distance from the hot surface. The temperature, T, is presumed to decrease linearly; the equilibrium partial pressure, $P°$, therefore decreases exponentially according to the Clausius-Clapyron equation. The actual partial pressure, P, is presumed to decrease linearly; hence, the supersaturation ratio, P/P_o, increases with distance from the hot surface. At some point a critical supersaturation is reached at which nucleation occurs. Soon after nucleation begins, nucleation and growth by condensation are complete and further growth occurs only by coagulation, (unless there are other substances such as sodium or sodium oxide present which will condense at a lower temperature).

The model shown indicates that if supersaturation results from supercooling a concentrated vapor then the parameters which determine the supersaturation may be: (1) the temperatures of the vaporizing surface, (2) the temperature of the sweep gas and (3) the velocity of the sweep gas (which determines the thickness of the stagnant layer).

Figure 5 shows a model of a situation in which supersaturation results from a gas-phase chemical reaction. At the top is shown a diagram which indicates a more-or-less stagnant mixing zone. One side of the mixing zone is swept by a velocity of nitrogen containing oxygen. The graph indicates schematically that the sodium concentration decreases from a high value at the left side of the zone to zero at the other. The oxygen concentration increases from left to right. The concentration of sodium oxide product will be a function of the oxygen and sodium concentrations according to the reaction kinetics; it will presumably be a maximum near the middle of the zone. The equilibrium concentration of the oxide product may increase slightly toward the middle of the zone due to the heat of reaction. The supersaturation ratio would increase, schematically as shown, until the critical value was reached above which nucleation would occur.

If the supersaturation results from a gas-phase reaction; it may therefore depend on: (1) the concentration of reacting species, (2) the temperature of the carrier gases, and (3) the rate of mixing or turbulence.

Homogeneous nucleation has been considered in order to suggest the parameters which might determine primary particle size. It has been indicated that primary particle size may depend on surface tension, temperature, radiation and supersaturation. Supersaturation, in turn, may depend on other parameters which have been noted.

Early Growth by Coagulation

Another topic is now introduced. We may ask now, what early growth by coagulation does to the particle size distribution. It is possible that after only a short period of coagulation, virtually no primaries remain and the size distribution no longer depends on the primary particle size. Spiegler and coworkers(5) considered the possibility some time ago; their results are shown on

Fig. 6. This is a plot of normalized volume fraction distribution vs particle radius. The curves labeled t = 0 were asserted initial size distributions; in other words, they represent two grossly different guesses of the primary particle size distribution. The curves labeled $t = 10^{-3}$ sec etc. are the calculated distributions after thermal coagulation for the times indicated. As you see the two distributions come together, they are close at 10^{-2} sec and are indistinguishable after 10^{-1} sec.

Although this calculation was not specifically done for UO_2, it applies to that case. The value of Ø, the volume fraction (i.e., the volume of particle phase per unit volume of aerosol) was taken to be 3.7×10^{-7} in this calculation. The concentration of saturated UO_2 vapor at the melting point of UO_2 corresponds to about 8×10^{-7}. In other words, the value of Ø used in this calculation is of the same order as one would expect in the nucleation zone near molten UO_2.

So if the aerosol is not diluted within times in the order of milliseconds, coagulation will probably erase the possible gross effects of primary particle size. It seems reasonable that under some conditions, significant dilution will occur in such short times. So, if the nucleation environment is such that dilution occurs quickly, the size distribution of the agglomerates may depend on primary particle size, otherwise rapid coagulation will obliterate this effect of the primary particle size.

(I should note also that the results of J. R. D. Stoute and J. F. Van de Vate as reported at this conference are pertinent to these considerations; they indicate that the settling rates of similar sized agglomerate particles depend significantly on the size of the primary particle.)

Secondary Growth by Condensation

Figure 7 introduces another problem, that of secondary growth by condensation. If there is a large amount of vapor (say of sodium or sodium oxide or in the water-cooled reactor situation if there is a quantity of water vapor) then very large increments of secondary particle growth can occur. I would like simply to suggest two extreme situations in regard to secondary growth by condensation which would lead to two extremely different results. The first is shown on Fig. 7. If the aerosol is cooling rapidly so that the supersaturation is high, then, to a first approximation, the vapor molecules will each condense on the nearest nuclei surface. The results would be that every nucleus would grow by the same amount. On this graph is shown a plot of normalized number distribution vs particle radius. An initial, r^{-4} distribution between a minimum radius of 0.04 micron to a maximum radius of 0.7 micron was assumed. Three other distributions are indicated which would result from growth by condensation from three different vapor concentrations. Remember, the assumption was that every nucleus received an equal share of the condensing vapor. In the extreme case, with C_o (i.e., initial concentration of condensing vapor) equal to 100 $\mu g/cm^3$ an essentially monodisperse aerosol resulted. With lower condensing vapor concentrations the size distributions have significant widths.

Figure 8 suggests the other extreme. If condensation of the secondary vapor occurs at low supersaturation, it may happen, as a consequence of the relationship expressed by the Kelvin equation (which was explained earlier), that only the larger particles in the size spectrum will grow. In principle, a sharp cutoff could occur; all particles smaller than the cutoff would not grow and all larger would grow. The approximate effect on the number concentration distribution is shown on Fig. 8. The result would be that the tiny particles would be left unchanged; there would be a gap in the size distribution (i.e., a range of particle radii in which there were no particles) and then there would be a distribution of some larger particles. Existing data(6) suggest that under some conditions of growth of UO_2 agglomerates in condensing sodium, this cutoff growth phenomenon may have occurred.

In summary, growth by coagulation, near the nucleation zone, may quickly achieve an agglomerate particle size distribution which is independent of the primary particle size. (The work of Stoute and Van de Vate, reported at this conference, indicates, however, that the settling behavior of the agglomerates depends significantly on primary size.) Secondary condensation, if it occurs at high supersaturation, may cause all nuclei to grow equally. This would lead to an efficient washout mechanism. Secondary condensation at low supersaturation could leave a bothersome aerosol of the tiniest nuclei.

Suggested Experiments

Figure 9 suggests an experiment to study nucleation and growth by condensation. To study nucleation, there would be no preformed nuclei in the gas stream. Nucleation would then occur homogeneously in this experiment due to supersaturation which, in turn, would be due to supercooling a concentrated vapor. The parameters (which are temperature of the vaporizing surface, temperature of the gas stream and velocity of the gas) would presumably describe the nucleation zone, as explained earlier.

One would measure primary particle size to see if it could be affected by the controlled parameters.

To study secondary growth by condensation in the same experimental setup, one would put an aerosol into the gas stream of various concentrations and size distributions. One would measure size distribution of the effluent aerosol.

Figure 10 schematically shows another suggested experiment. Here one would generate supersaturation by means of a gas-phase chemical reaction. Homogeneous nucleation could be studied (without preformed nuclei in the gas stream). The temperatures and concentrations would be controlled. Presumably the turbulence in the mixing zone could be varied by changing the size of the orifice. Primary particle size would be measured vs controlled parameters.

Summary

In summary, I think that nucleation is probably not the most important process, i.e., nucleation probably does not have as large an effect on eventual particle sizes as does growth. However, it is difficult to be certain of this, so some experiments on nucleation should probably be done. The purpose would be to see if the nucleation environment can be altered to produce significantly different primary particle sizes or significantly different size distributions.

Secondary growth by condensation (of sodium or sodium oxide in an LMFBR situation or of water in a water-cooled reactor situation) surely will, in many cases, substantially determine the particle size distribution. We need to be able to predict size distribution in order to estimate deposition rates, hence secondary growth by condensation must be thoroughly studied.

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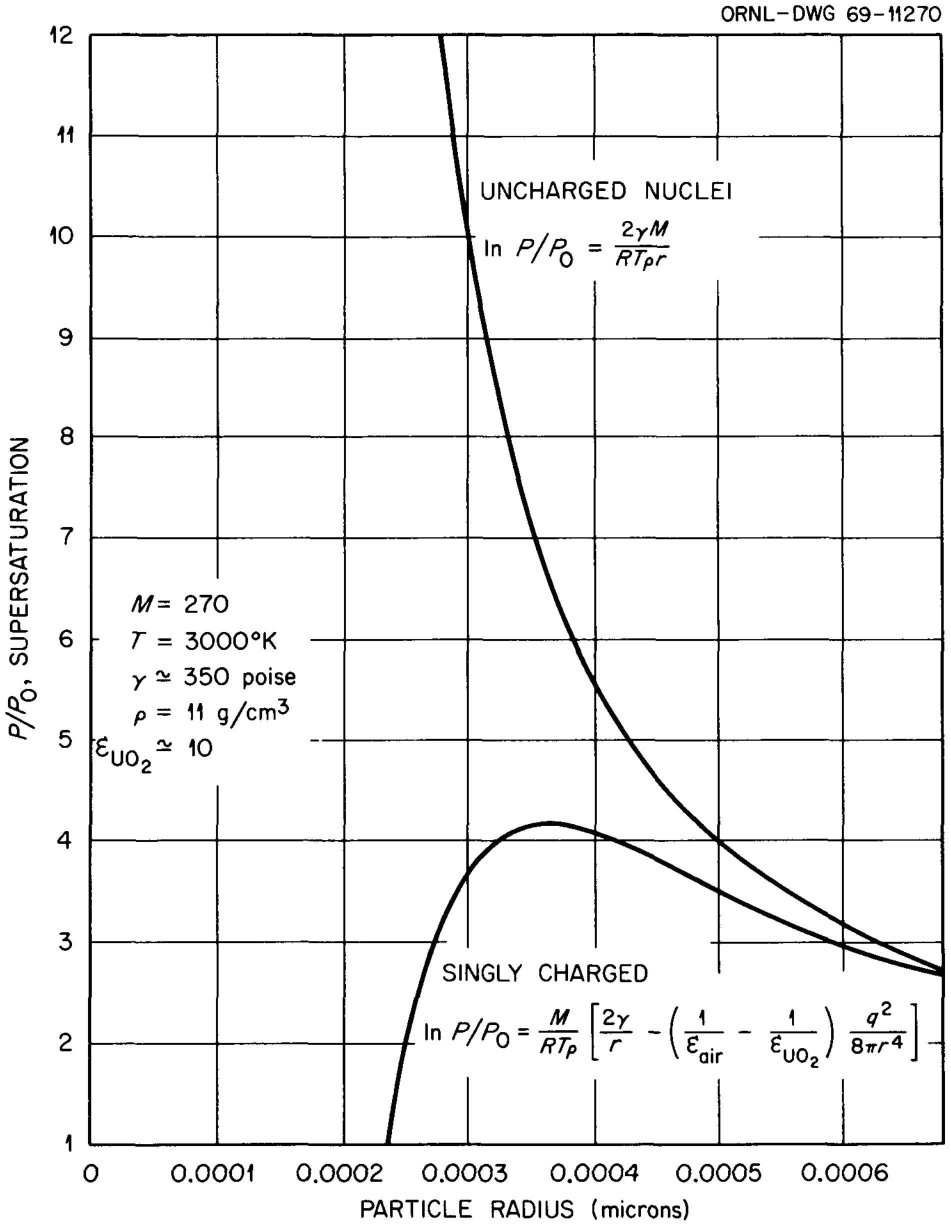
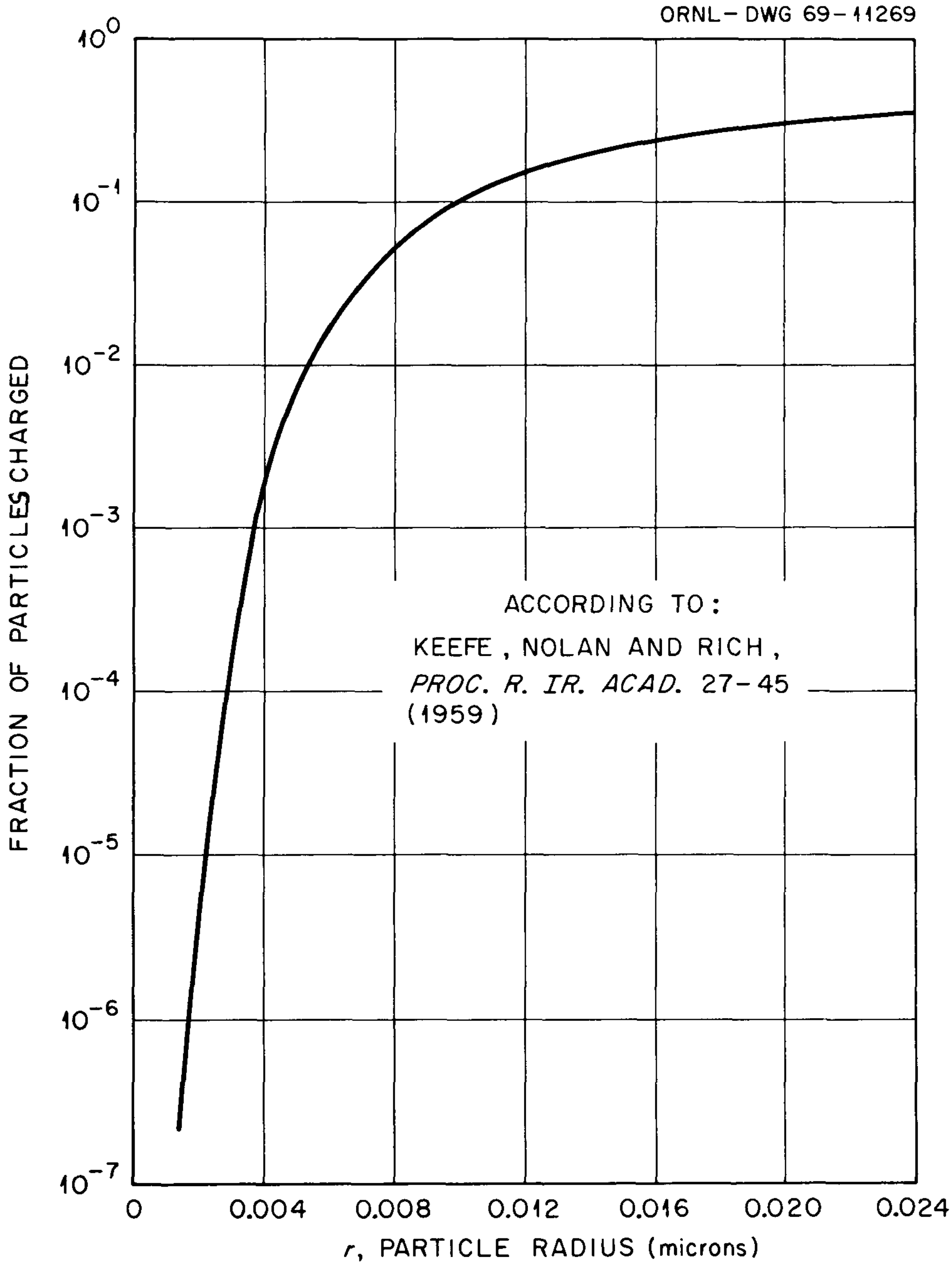


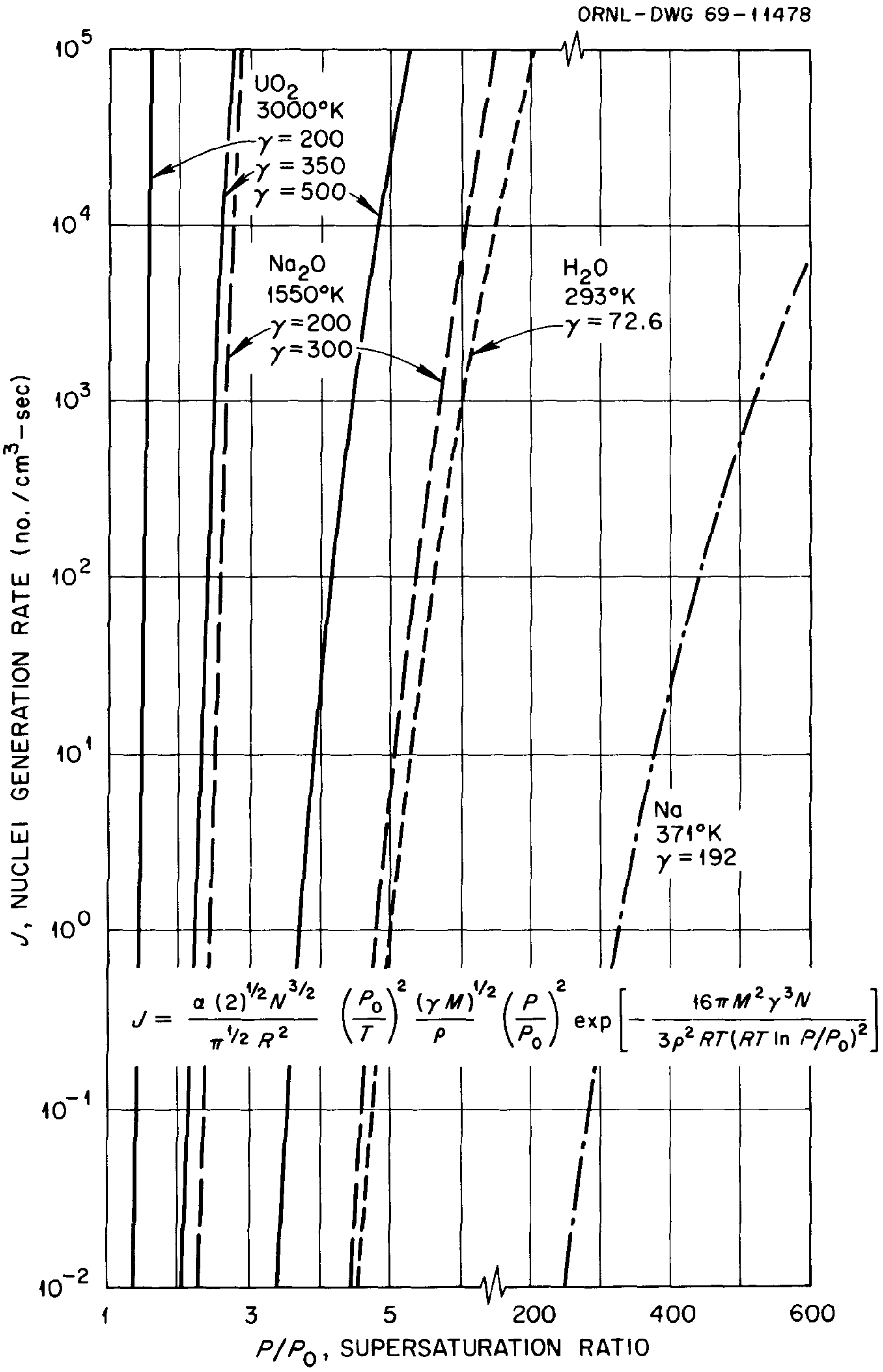
Estimated Equilibrium UO_2 Nuclei Radii vs Supersaturation.

Figure 1.



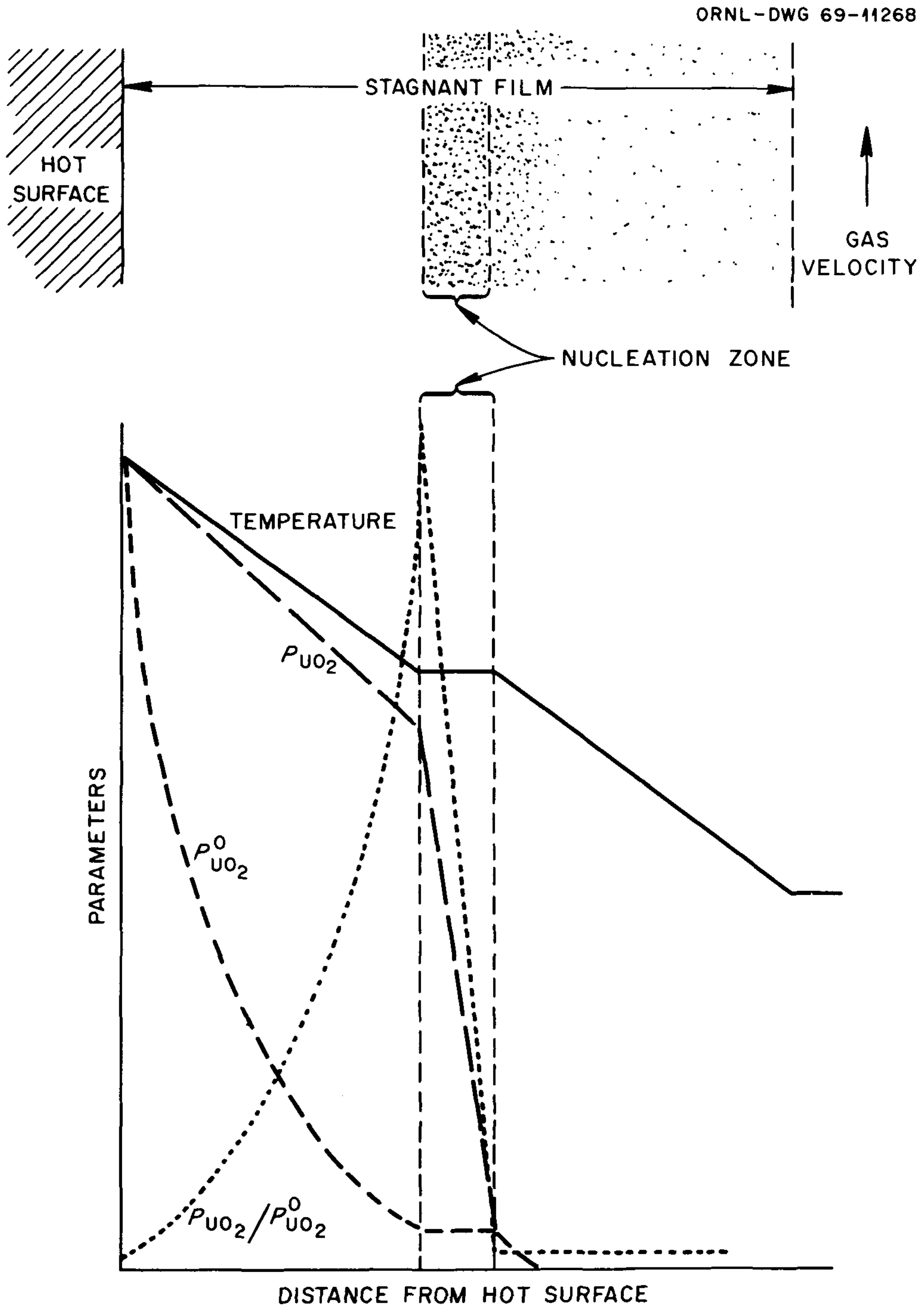
Fraction Charged of Particles in Equilibrium with a Bipolar Ion Atmosphere.

Figure 2.



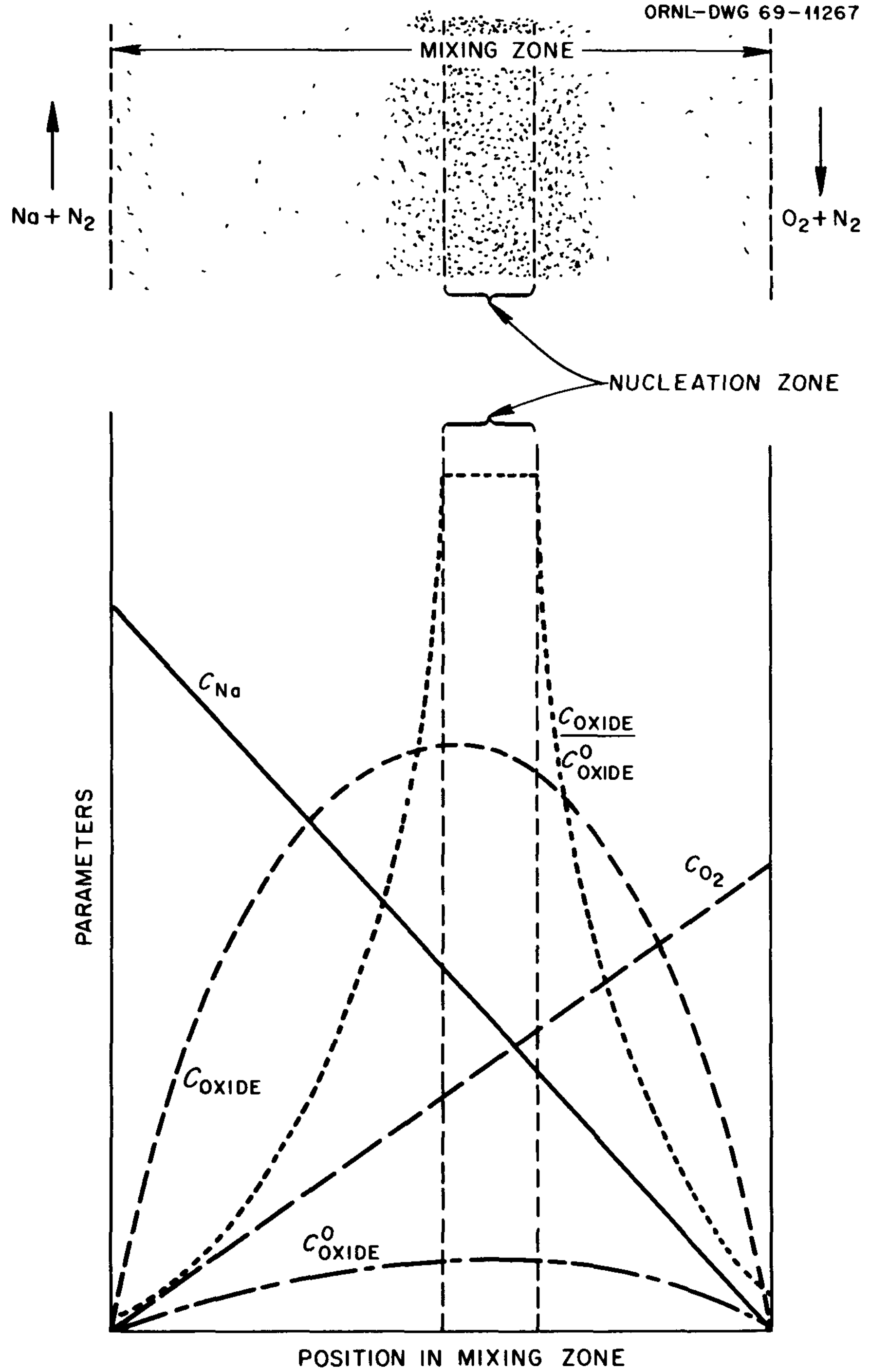
Estimated Nuclei Generation Rate

Figure 3.



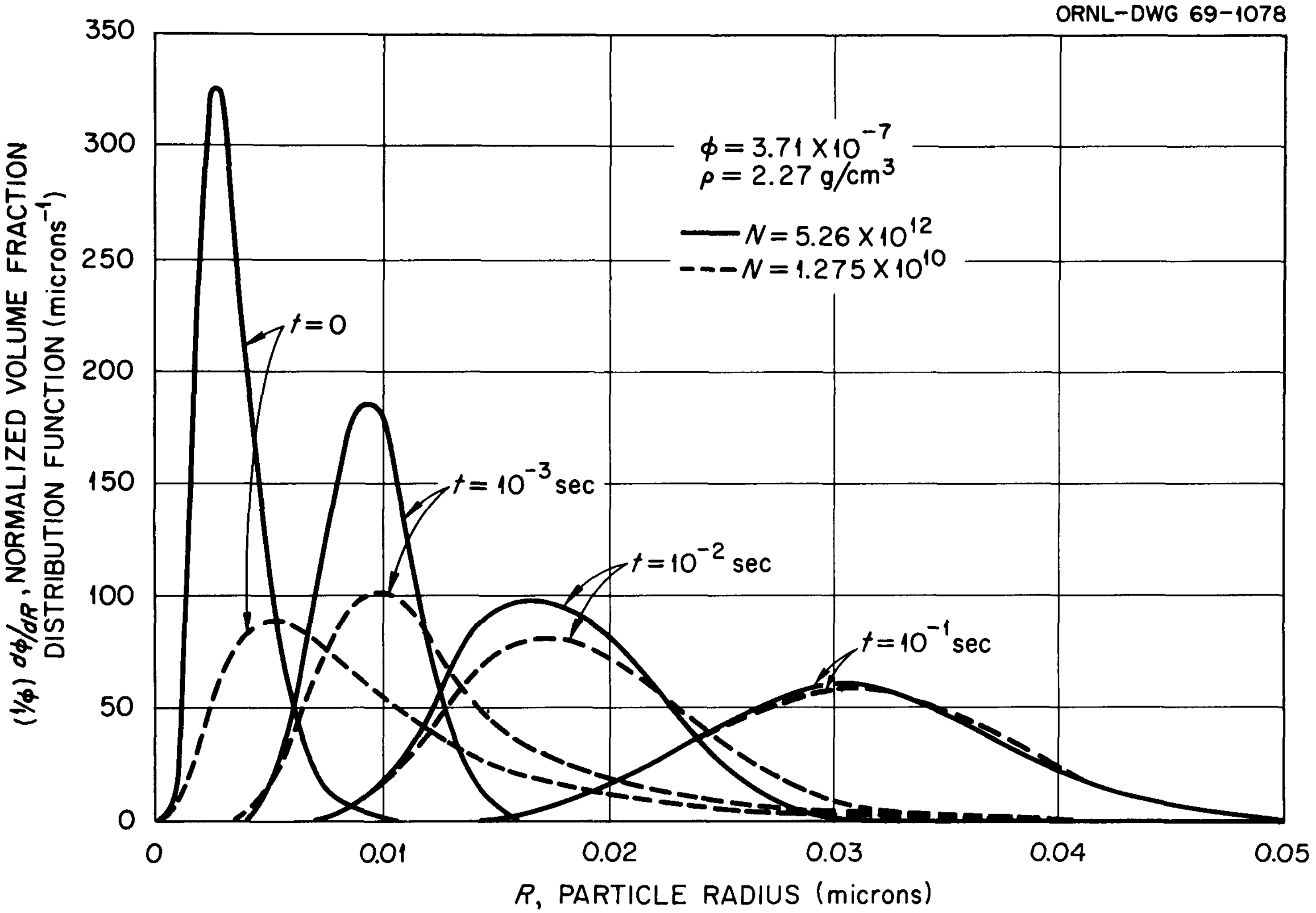
Model for Nucleation in Vapor Formed by Vaporization.

Figure 4.



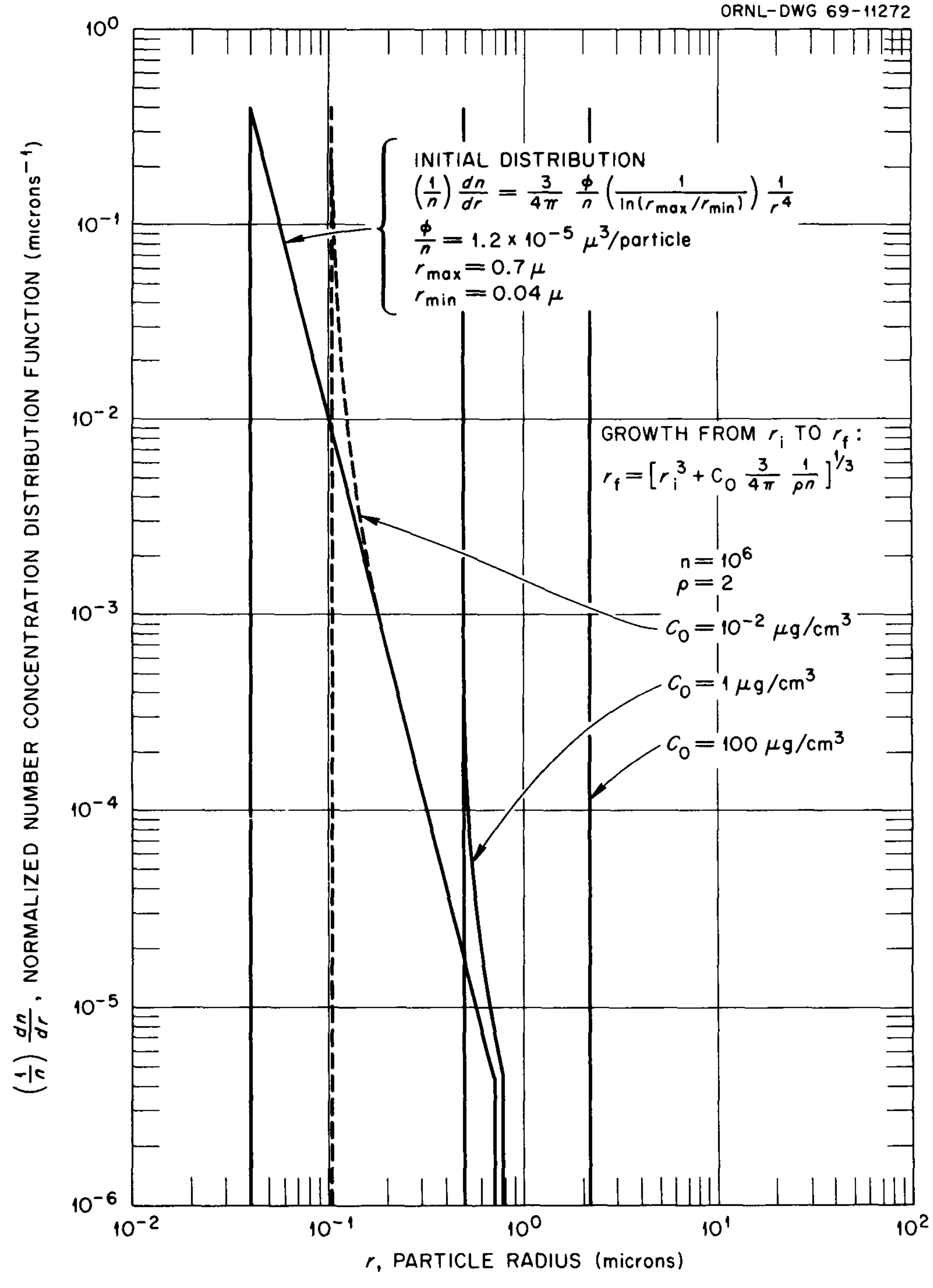
Model for Nucleation in Vapor Formed by Gas Phase Reaction.

Figure 5.



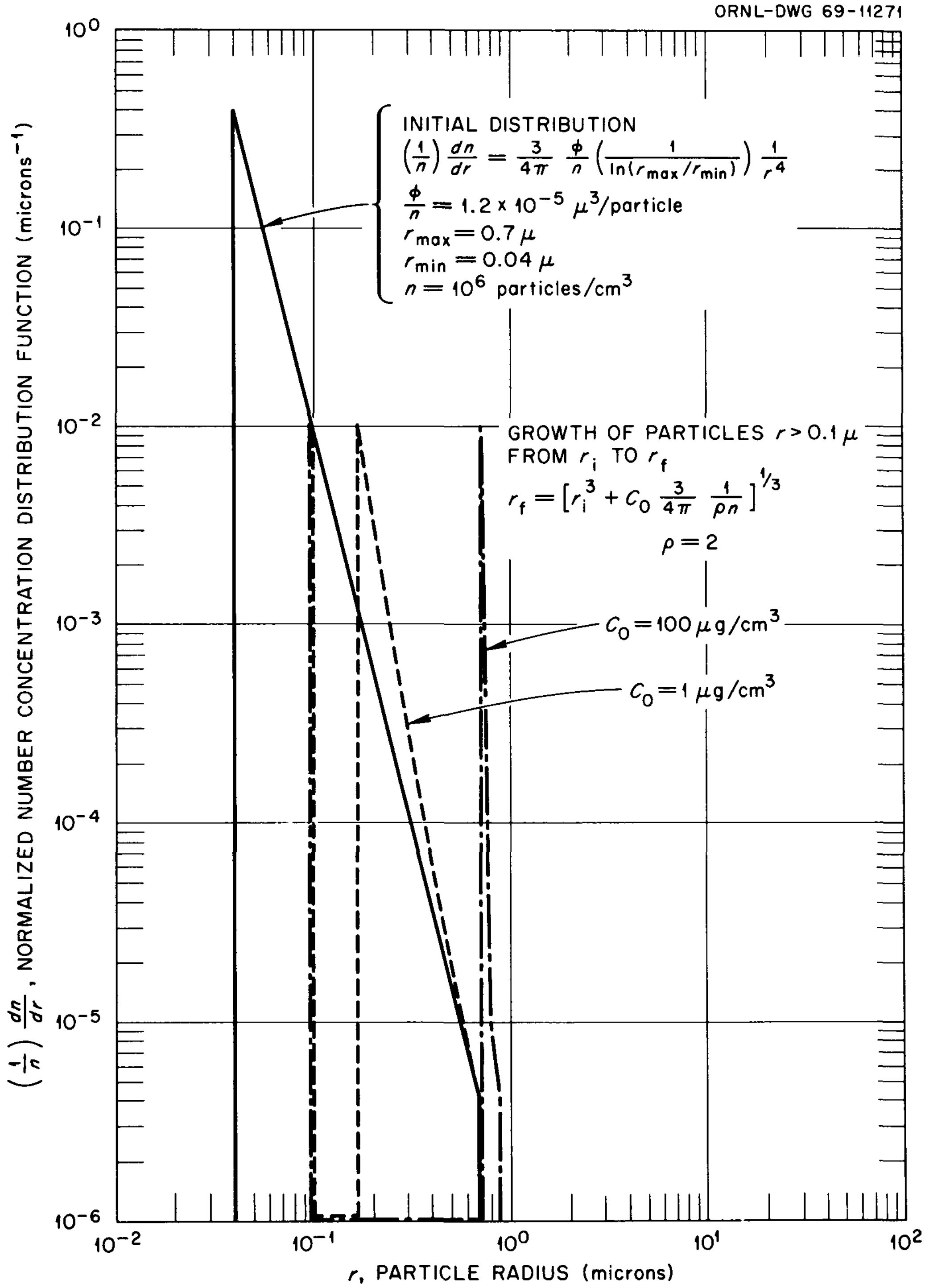
Evidence of a Steady-State Particle Size Distribution (Spiegler, *et. al.* NAA-SR-11997).

Figure 6.



Growth by Condensation at High Supersaturation.

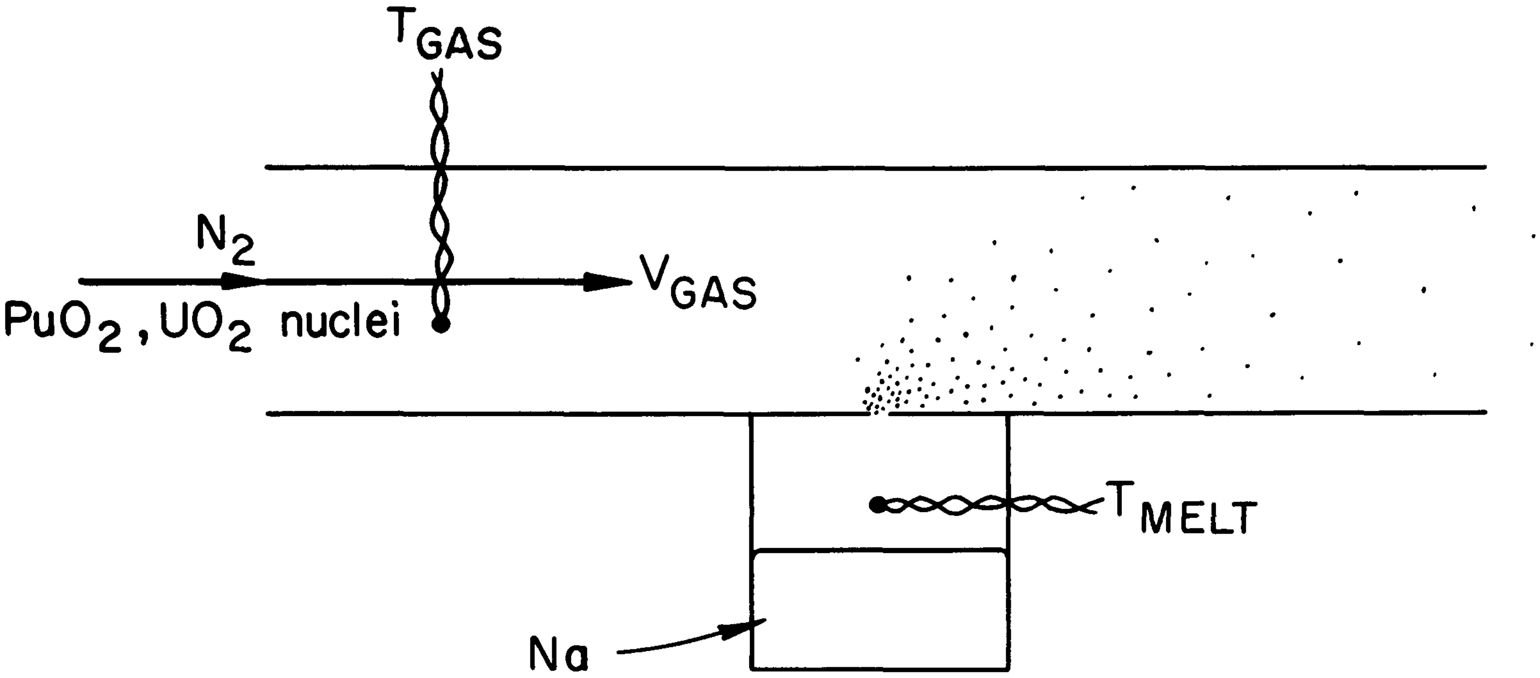
Figure 7.



Growth by Condensation at Low Supersaturation.

Figure 8.

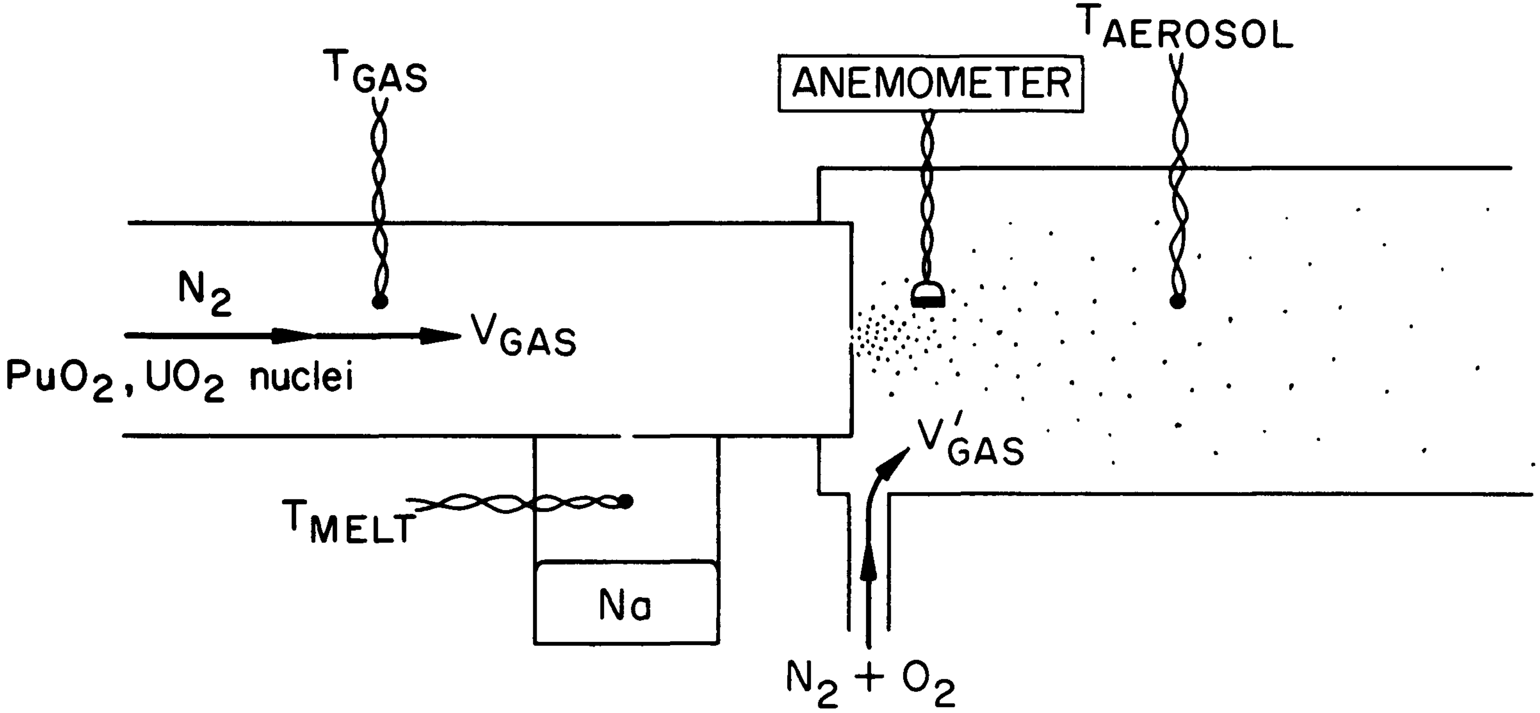
ORNL-DWG 68-6193R



Apparatus to Measure Primary Particle Size of Vapor-Condensation Aerosol.

Figure 9.

ORNL-DWG 68-6192R



Apparatus to Measure Primary Particle Size of Chemical Reaction – Condensation Aerosol.

Figure 10.

Paper Nr. 4

Contribution of the R.C.N. to the "Specialist Meeting on the behaviour of nuclear aerosols in closed systems".
(Karlsruhe, November 11th and 12th, 1969)
by J.R.D. Stoute and J.F. van de Vate

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1. Influences of formation on the characteristics and behaviour of aerosols.

1.1. Influence of specific formation energy.

This influence is studied by means of aerosols of identical initial mass concentration (3.5 mg/m^3) in the range between 4 kJ/g and 275 kJ/g and is not masked by any other effect such as mass concentration or tank dimensions.

Fig. 1 shows that the primary particles formed immediately after the explosive vaporization of the wire material, have a particle size distribution dependent on the specific energy dissipated during the explosion (Espec). The primary particles have a logarithmic-normal size distribution with a geometric median diameter (D_{50}) decreasing with increasing Espec. The standard deviation is between 1.4 and 1.6 and shows no relation with Espec. The primary particles are defined and measured as the particles forming the coagulates. They are analyzed by means of a semi-automatic particle size analyzer.

All decay curves of the particle concentration for different Espec show the same behaviour and lay in one band of experimental error (Fig. 2). The non-exponential initial decay of the particle concentration during the first 15 hours is attributed to coagulation accompanied with some deposition process. Therefore the decay is exponential, meaning that the polydisperse aerosol is present in an aerodynamic steady-state. This is in accordance with the view layed down in the R.C.N.-paper presented at the congress in Saclay (Nov. 4th to 6th, 1969). It means also that after the initial period the particle concentration behaves independent of the way the aerosol is formed and that it is only influenced by the tank dimensions.

The decay of the mass concentration is clearly influenced by the specific formation energy (Fig. 3). It can be seen that the initial decay rate is decreased by increasing the Espec. Afterwards the decay rate for the aerosols of the large Espec increases whereas at small Espec the effect is opposite. This suggests that the decay rate of the mass concentration approaches a certain value that appears to be roughly equal to the decay rate of the particle concentration.

Since plate-out enhances the decay rate of the mass concentration of a polydisperse aerosol, plate-out must have obtained an increasing importance with increased Espec. This will probably be due to the correlation between the primary particle size and Espec leading to a changed Fluffiness of the coagulates.

In case of a gold aerosol with Espec = 20 kJ/g sedimentation is the dominant mechanism upto 100 hours. This can be concluded from Table I: the ratio of half-life and height of the tank is roughly equal. The same table shows that this is not true for aerosols produced with Espec = 275 kJ/g. This was to be expected since we concluded earlier (from the course of the mass concentration) that for these aerosols plate-out plays an important role. (At the moment of editoring this report of the meeting experiments are finished by R.C.N. showing sedimentation to be negligible in case of Espec = 275 kJ/g. The half-life of the particle concentration is then proportional to the volume to surface ratio).

1.2. Influence of initial concentration of the aerosol.

The experiments ERGO 102 and 108 represented in Figs. 2 and 3 differ only in initial concentration. In both cases the specific formation energy amounts to 20 kJ/g.

The slopes of the mass concentration curves are strikingly different. The higher the initial concentration the steeper is the slope of the decay curve. Due to this at a certain moment the mass concentration of different experiments become equal and the curve of the larger initial concentration even crosses the curve of smaller initial concentration. Obviously coagulation is strongly increased in the early stages. When the coagulation has become negligible, the behaviour of the aerosol then formed, depends on its aerodynamic properties inherent in the primary particle characteristics and influenced by the coagulation. Hence as a large initial concentration will lead to an incrased mean particle mass by coagulation, sedimentation can play a larger role.

2. Mixed aerosols.

The behaviour of mixed aerosols with minority concentrations upto 10 wt% is idential with the pure aerosol of the majority component. The concentration of the additive remains constant during the aerosol life. This could be concluded from the following experiments:

- Na + SrO aerosols during the first 6 hours of the experiments;
- Au + 3 W% Cu aerosols in a time region of 60 hours.

TABLE I

R.C.N.-results about decay of number concentration at different Espec done in ERGO 200-(turnable)-tank

	Espec = 20 kJ/g		Espec = 275 kJ/g	
Tank height	$T_{\frac{1}{2}}$	$T_{\frac{1}{2}}$/height	$T_{\frac{1}{2}}$	$T_{\frac{1}{2}}$/height
2 m	18.4 h	9.2 h/m	29 h	14.5 h/m
1 m	10.2 h	10.2 h/m	-- h	-- h/m
0.5 m	4.7 h	9.4 h/m	19 h	38 h/m

Technical data for experiments of ERGO 100 and 200 series

	Volume	Height
ERGO 100	1.2 m^3	1.50 m
ERGO 200	1.0 "	2.0 "
	1.0 "	1.0 "
	1.0 "	0.5 "

Sampling and analysis:

- Mass concentration: by loading Sartorius membrane filters with aerosol labeled with ^{198}Au-tracer. Mass determination by comparison of count rates of filter loads with a standard of same specific activity.
- Particle concentration: by counting the particles on a known area of an electron micrograph of the grid loaded with particles from a knwon aerosol volume by electrostatic precipitation.

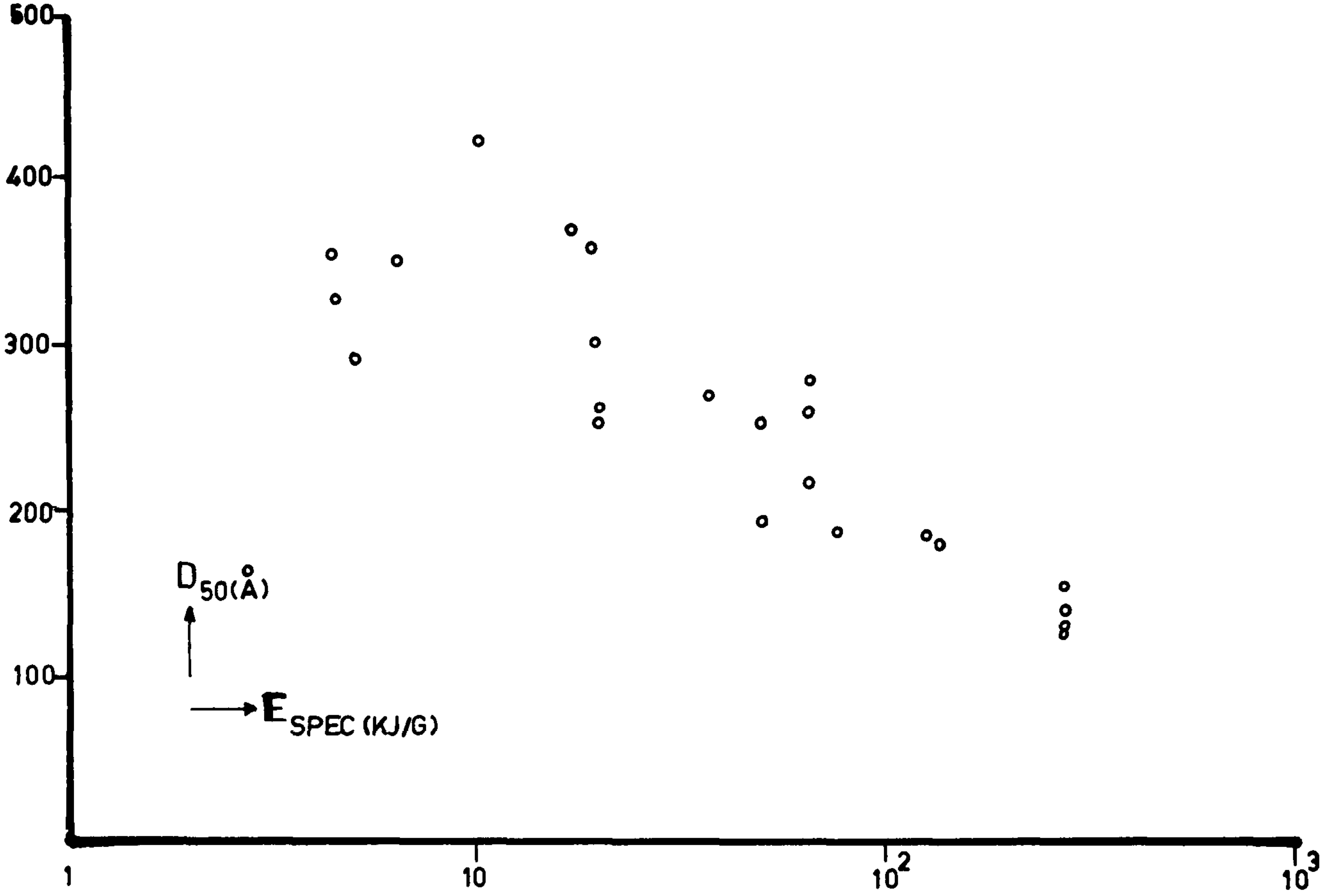


Fig 1 Geometric median diameter (D_{50}) of primary gold particles formed by explosion of gold wires with different specific energies.

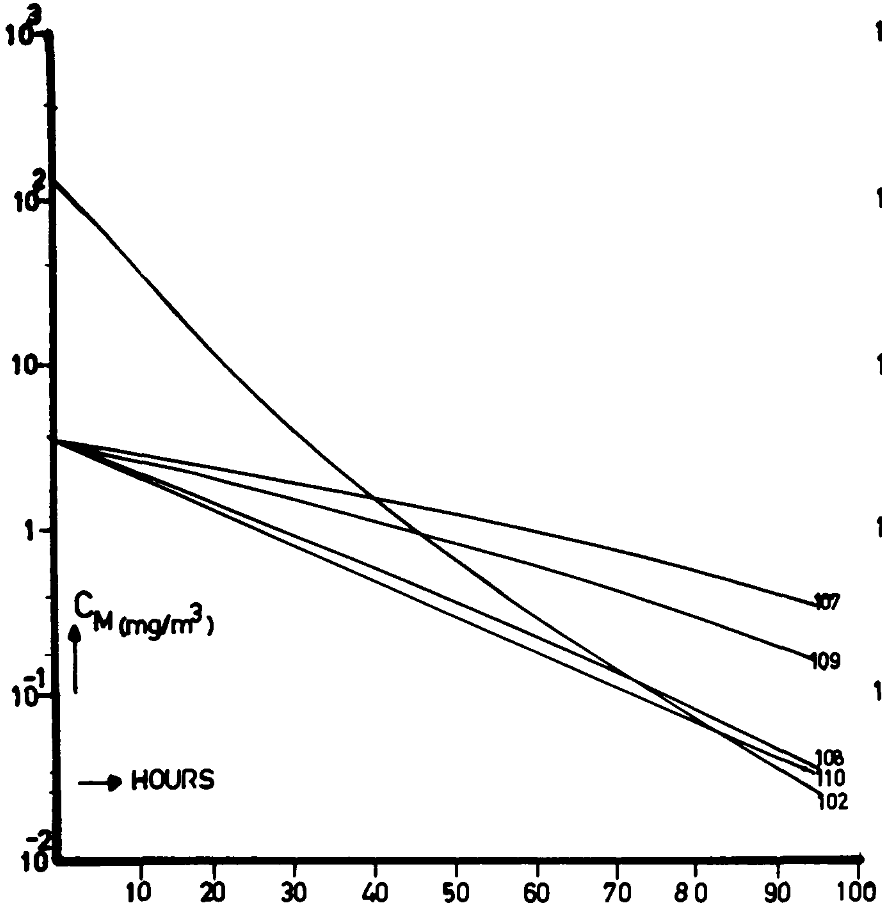
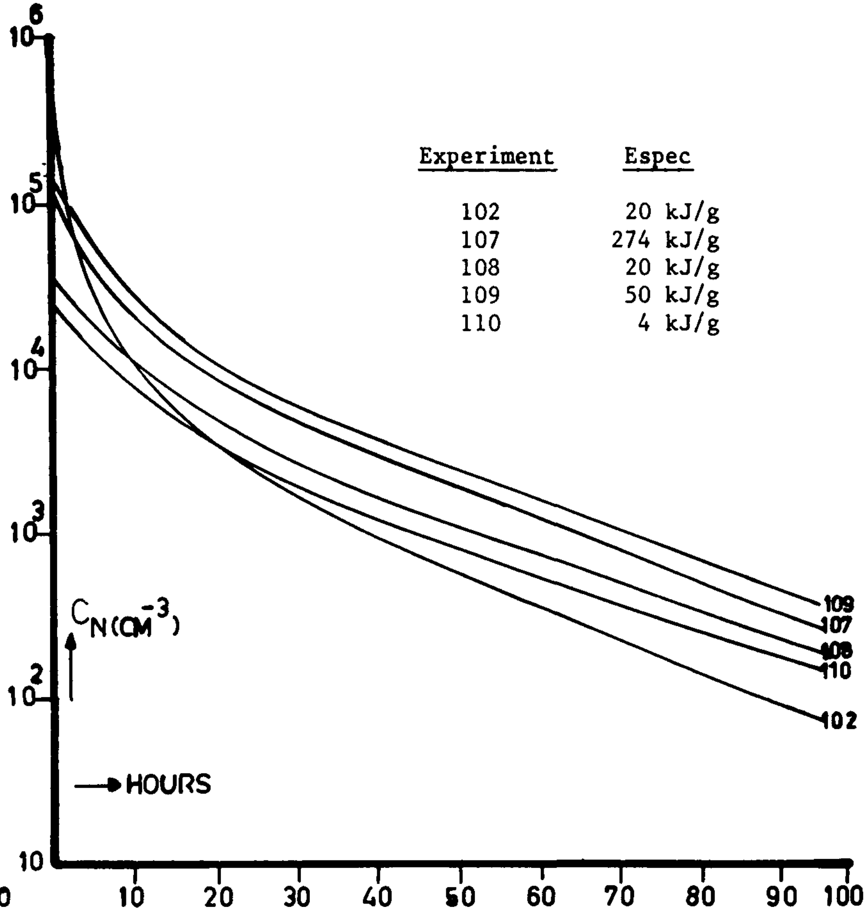


Fig. 3 Mass concentration as a function of time for gold aerosols generated with different specific energies (see also insert Fig. 2).



Experiment	Espec
102	20 kJ/g
107	274 kJ/g
108	20 kJ/g
109	50 kJ/g
110	4 kJ/g

Fig. 2 Particle number concentration as a function of time for gold aerosols generated with different specific energies.

Some recent results on the behaviour of nuclear aerosols observed in TUNA

von

H. Wild
Institut für Angewandte Reaktorphysik
Kernforschungszentrum Karlsruhe

1. Introduction

In the scope of the Karlsruhe research program on nuclear aerosols [1] series of experiments with UO_2-aerosols has been carried out in TUNA which may contribute to the understanding of the behaviour of nuclear aerosols.

The generation of aerosols has been accomplished by means of the exploding wire technique. The energy inserted into the UO_2-probe has been ca. 500 Wsec/g, the insertion time has been varied between 0,1 msec and 3 msec depending mainly on the inductivity of the electric circuit. The two different free volumes chosen for the aerosol experiments have been: the main vessel of 2.2 m^3 and with a hight of 2.90 m and the aerosol chamber of 0.02 m^3 and with a hight of 0.3 m. There was no change in the generation conditions of the UO_2 aerosols for both volumes.

This paper discusses the time function of the particle number concentration of the UO_2 aerosols and its relation to aerosol processes. More results obtained in the experiments on the particle size distribution and mass concentration are reported in [2] in greater detail.

2. Experimental results

In figure 1 and 2 typical particle number concentrations as function of time are given which we have measured by use of a condensation nuclei counter in TUNA.

Three time ranges can be identified out of our experimental results:

- range 1 with the fast decrease of the particle number concentration
- range 2, a transition range with a much slower decrease than range 1, and
- range 3, with a once more slow and exponential decrease of the particle number concentration.

The question arises how these ranges can be correlated to specific aerosol processes and, therefore, made computable and predictable in other cases. Certainly, the first range is controlled by the coagulation process. The high concentration of particles must lead to frequent contacts due to diffusion of particles resulting in coagulation. This is verified by the application of the simple coagulation law of Smoluchowski [3]

$$\frac{1}{c_p} = \frac{1}{c_{p0}} + Kt \qquad (1)$$

The second range may be a transition range of nearly one day, in which the concentrations proceed into the range three.

The third range follows with very good agreement an exponential function. This may indicate that the exponential law of sedimentation (stirred settling) [4] governs this time range according to

$$c_p(t) = c_{p0}\, e^{-\gamma t} \qquad (2)$$

with

$$\gamma = \frac{1}{18} \cdot \frac{g}{\eta} \cdot \frac{\rho d_6^2}{H}$$

It denotes

c_p = particle number concentration
c_{p0} = initial concentration without sedimentation
g = acceleration due to gravity
η = viscosity
ρ = particle density
d_6 = geometric mean weight diameter
H = height of free volume
t = time

It should be noted that the agreement with an exponential law is not a sufficient argument for the assumption of the sedimentation process only. The decrease of the particle number concentration due to diffusion losses can also be exponential. But it is conceivable **that** diffusion losses may not be dominant because of the relatively large particle sizes in the range of 0.3 µm, which we have found /‾2_7. This may be illustrated by a simple consideration on the average time necessary to remove a particle from the aerosol population by the diffusion process or by the sedimentation process. Einstein's correlation /‾4_7 of the mean square displacement of a particle by thermal motion (or diffusion) can be written in the form

$$t_{Diff} = \frac{R^2}{2D} \tag{4}$$

Introducing the corresponding values of our experiments R = vessel radius = 50 cm and $D = 3 \cdot 10^{-6} \frac{cm^2}{sec}$ for spherical particles of a diameter of 0.3 µ /‾4_7 the time t_{Diff} appears to be in the order of 70 years. This means that the diffusion process works fairly slow compared with the time scale in our experiments. If we consider the sedimentation process a similar time can be derived, namely

$$t_{sed} = \frac{H}{v_{sed}} \tag{5}$$

with the vessel height of 324 cm and a sedimentation velocity of about

$5 \cdot 10^{-4}$ cm/sec for particles of 0.3 μm diameter the parameter t_{sed} results to 7,5 days. This indicates that the sedimentation process is probably of much greater importance than the Browns'an diffusion.

In conclusion of these facts we assumed that coagulation and sedimentation alone might already give a good explanation of measured time functions. Therefore we made the attempt to explain the behaviour of the particle number concentration by a semiempirical equation including coagulation and stirred settling.

3. Semiempirical equation for the particle number concentration

In trying to develop a correlation or equation which describes the time behaviour of the particle number concentration we made the assumption that the superposition of the mathematical expressions of these both processes (coagulation and stirred settling) might give already a consistent description of the time function. Of course, this assumption implies three bounding conditions:

- no influence of the particle size and its change with time
- no influence of additional processes in all three time ranges
- no influence of the two processes against each other.

Therefore, as one would axpect, the simple additive correlation of the equations 1 and 2 did not give a good agreement with the experimental curves. However, we were successfull by inserting a corrective term and forming a time dependent coagulation constant according to

$$K = K_o e^{\alpha t} \qquad (6)$$

Utilising equations (1), (2) and (6) one can write the following equation for the particle number concentration as function of time

$$c_p(t) = \frac{c_{p0}}{1+c_{p0} K_0 t e^{\alpha t}} + c_p(t_s) e^{-\gamma(t - t_s)} \qquad (7)$$

where t_s is the time when only sedimentation starts to be effective. This expression is in good agreement with the experimental curves (fig. 1 and 2) if an empirically determined α is used. The parameter α changes with the experiment volume and was determined to be for the main vessel experiment
$\alpha_V = 0{,}13 \cdot 10^{-4}$ sec^{-1} and for the aerosol chamber experiment
$\alpha_A = 0{,}91 \cdot 10^{-4}$ sec^{-1}.

Literature

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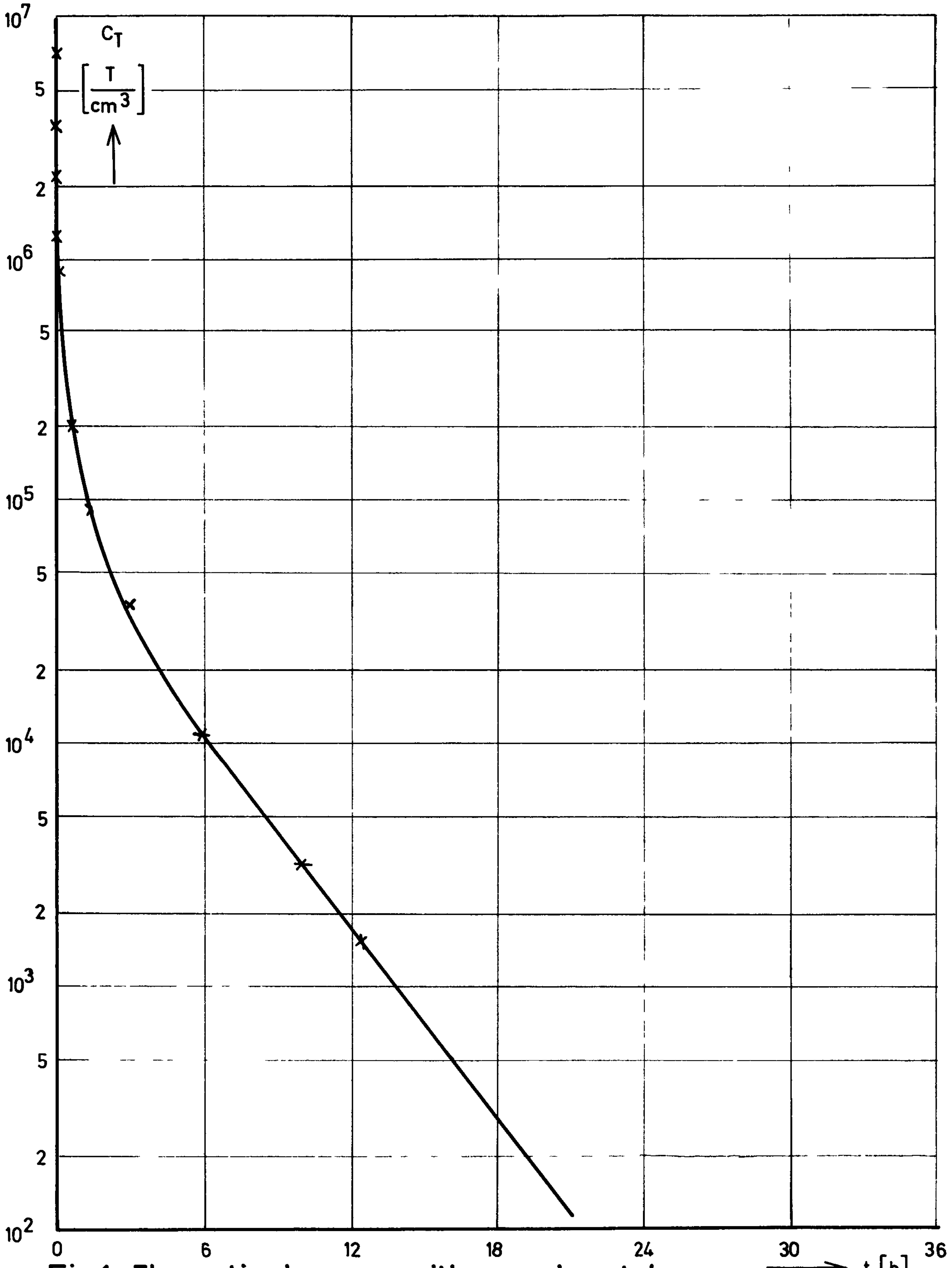

Fig.1 Theoretical curve with experimental points (x) in the aerosol chamber (163)

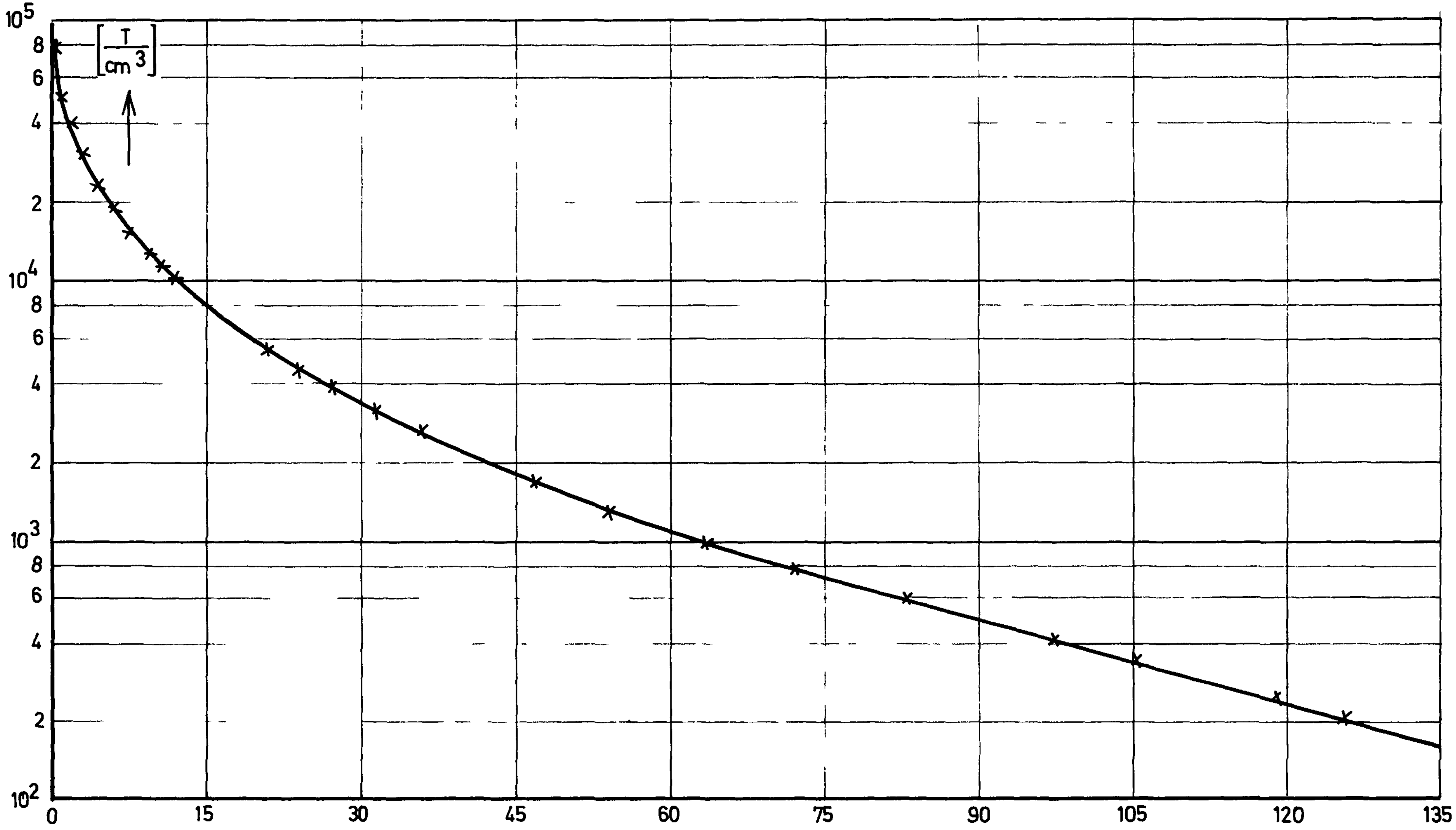

Fig. 2 Theoretical curve with experimental points (x) in the TUNA (166)

JAERI Research Program on the Behavior of Nuclear Aerosol and Radioiodine in an Accident of the Fast Sodium Reactor*

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I. Introduction

Fast sodium-cooled plutonium reactors deserves careful consideration in the safety aspects. In the hypothetical major accident, vaporized fuel mixture, sodium and fission products are dispersed in the primary reactor container by the explosive force. In order to estimate the radioactivity of accidental release and assess the hazard to surrounding population, the JAERI program to study the behavior of nuclear aerosol in the reactor container was started.

The principal objective of program is to have an insight of the parameters, which are involved in the evaluation of hazards. The mass concentration of plutonium aerosol and its decrease rate in the reactor container may vary with mass concentration, particle-size distribution, thermal condition and container shape.

II. An outline of the program

The experimental vessel is of stainless steel—a cylinder, 80 cm in diameter and 2 m high. Uranium oxides and other metals

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* The program is carried out under the contract with Power Reactor and Nuclear Fuel Development Corporation.

will be vaporized in the vessel by means of an arc generator, whose power is variable up to 20 kw. Sodium aerosol is produced with a resistance heater.

The shape of plutonium and uranium oxide aerosols differ from those of other metals. The reason is not known well. By knowing the behavior of several kinds of aerosol such as sodium, lead, zinc and magnesium, the characters of nuclear aerosols can be known. The mass concentration, wall plating and floor deposition of airborn materials will be measured by quantitative analysis with absorption spectrophotometer or fluorescence spectrophotometer.

It is assumed by the collision theory that the higher the initial concentration the larger the rate of decrease in the concentration of the aerosol; this decrease in concentration is very rapid. It is therefore desirable to follow the change of concentration "continuously". An instrument can be used for this purpose, which consists of an electric device to measure the intensity of light scattered by the aerosol. The original instrument was developed by Koshi$^{1)}$. The instrument and the counting system were modified for our purpose by JAERI. The following equation was derived by Koshi to express the response R of the instrument.

$$R = K\sigma \frac{C}{P} \exp\left(-\theta \frac{C}{P}\right)$$

where K is a constant, σ the relative scattering coefficient of aerosol, P the dissipation coefficient of aerosol particles, θ the linear function of the extinction coefficient of particles and C the mass concentration of aerosol.

On the other hand, if the temperature of the wall of the vessel is lower than that of the contained gas, the gas may flow to the wall, promoting the rate of plating of airborn particles onto the wall. This condition may exist in certain types of reactor container. This thermal effect may be investigated by controlling the wall temperature with a heater and a heat-sink arround the wall.

There are also other items of research in the program, in addition to the one described outlined above:

(a) to investigate the behavior of airborn particles with monodisperse aerosol of liquid and solid material,

(b) to investigate the behavior of fission iodine in a sodium fire,

(c) to develop a method of controlling the sodium aerosol,

(d) to develop a method for rapid decrease of the concentration of plutonium aerosol in accidental condition.

Reference

1) Shigezi Koshi: "On the characteristics of the digital dust indicator", Ind. Health, 2, 189 (1964)

7. Conclusions

At the end of the specialist meeting on the behaviour of nuclear aerosols in Karlsruhe (Nov. 11 - 12, 1969) a final discussion took place on the conclusions to be drawn from the discussions and presentations held at the meeting. The following points appeared to be of significance from the standpoint of present knowledge and of future work on nuclear aerosols.

1. The theoretical understanding of the processes involved in the behaviour of nuclear aerosols in closed vessels has substantially increased during the recent time. In the concentration range of the present experiments, coagulation is the dominant mechanism for short times while sedimentation predominates after long aging-times. Sedimentation may or may not be accompanied by diffusion to the vessel walls, depending primarily on the geometry of the enclosure.

2. The most important geometrical parameter is the height, H, of the vessel under consideration. For stirred settling, a correlation of the ratio of H to the half-time can serve as a useful tool for estimating the time dependence of the aerosol particle number concentration during the long-term settling regime. Experimental verification of the reciprocal relationship between vessel height and half-time exists only up to H = 3m. The proportionality constant may depend on the surface to volume ratio.

3. The energy involved in the formation process influences the primary particle size distribution, hence the fate of the aerosol population. Higher inserted energy gives smaller mean geometrical particle size.

4. The influence of the aerosol formation on the time behaviour of mass- and particle concentrations is still unknown. A promising theory seems to indicate that based on the assumptions of an instantaneous source and the coagulation of spherical particles to form spherical agglomerates, an "initial" particle size distribution can be predicted. It is still subject of definition what the term "initial" means.

5. For higher primary mass concentrations the decrease in the first phase of time function of the concentration is faster.

6. The following recommendations for future work can be given:

- experiments should be carried out using adequate instruments to evaluate the weight of the different fundamental processes influencing the aerosol
- experiments with high initial mass concentration in the order of 1 g/m^3 and above should be carried out
- methods of generating high mass concentration aerosols and the influence of these various methods on the particle size distribution immediately after formation and on the long term should be studied
- experiments in larger vessels as well as in vessels with different surface/volume ratios (volume of 50 m^3 and above, height of 5 m and above) should be carried out with particular attention given to an investigation of the effect of varying vessel height on the time dependence of the aerosol mass concentration.
- mixed aerosols composed of sodium and fuel particles should be studied with particular emphasis placed on an investigation of the effect of the Na/fuel ratio.
- additional theoretical work is required on methods for treating the coagulation and settling of chains.
- both theoretical and experimental work is needed on methods to account for particle impaction on surfaces in the event of large convection currents which will undoubtedly exist in a real accident.
- additional experimental work is needed to develop suitable simulants for PuO_2, for use in large-scale tests. Comparison tests between the behaviour of these simulants and PuO_2 aerosols are needed.