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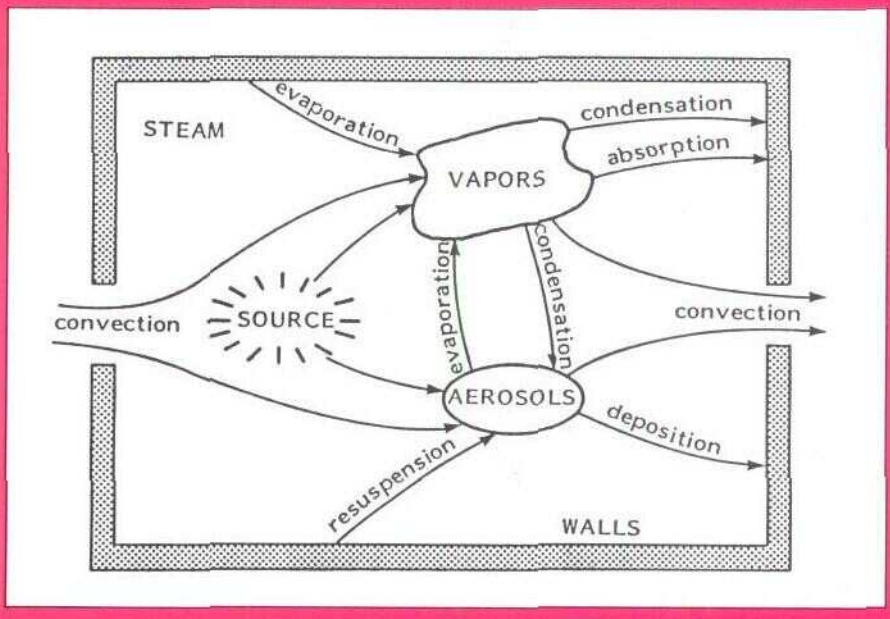

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AEROSOL TRANSPORT IN SEVERE REACTOR ACCIDENTS

Final Report of the NKA Project AKTI-160

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I ABSTRACT

Aerosol behaviour in the reactor containment was studied in the case of severe reactor accidents. The study was performed in a Nordic group during the years 1985 to 1988. Computer codes with different aerosol models were used for calculation of fission product transport and the results are compared. Experimental results from LACE, DEMONA and Marviken-V are compared with the calculations. The theory of aerosol nucleation and its influence on the fission product transport is discussed. The behaviour of hygroscopic aerosols is studied. The pool scrubbing models in the codes SPARC and SUPRA are reviewed and some calculational results are reported. The present status of knowledge in this field is assessed on the background of an international rewiew.

Keywords: Comparative evaluations;
Computer codes;
Containment systems;
Hygroscopicity;
Meltdown;
Nucleation;
Radioactive aerosols;
Radioactivity transport;
Reactor cores;
Reactor safety;
Scrubbing

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Finland: Technical Research Centre
Finish Centre for Radiation and Nuclear Safety
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II SUMMARY

Studies of a hypothetical severe accident in a nuclear power reactor includes the behaviour of aerosols that may be released from a molten reactor core. Aerosols can be described as small airborne particles. When released from a reactor core they are highly radioactive. Enclosed in the primary circuit or in the containment they will deposit within a time scale which is dependent on their size distribution and on the geometry of the compartment. The thermal-hydraulic and chemical conditions are also important. The size distribution changes with time due to agglomeration, deposition, chemical effects as hygroscopicity, and production of new particles.

The task for aerosol calculations is to predict the deposition and transport under the given conditions. The determination of the aerosol source is normally a task for chemists, but such calculations are also sometimes made in connection with transport calculations as they are strongly interdependent.

Particles are produced as long as the core debris has a sufficiently high temperature or if deposited particles are re-evaporated due to a high surface temperature or if mechanical effects cause resuspension. The aerosols which do not deposit will eventually leak out to the environment and cause radioactive contamination. It is therefore important to know the deposition rate and whether deposited aerosols are subject to resuspension or revaporization with risk of subsequent leakage to the environment.

Aerosols are mainly produced in the primary circuit as the temperature is highest in this region, especially in the reactor tank, but also in the reactor cooling system. Aerosol production starts at a temperature much below the melting temperature of the fuel. The main mechanism is then diffusion in gas form to the surface of the fuel where the gas leaks out to the surrounding atmosphere. Molten fuel will of course vaporize fission products and other constituents to a much higher rate. At some distance from the fuel the temperature is sufficiently low for condensation of the evaporated material. This occurs mainly as nucleation in the atmosphere whereby aerosols are generated.

The size distribution of the particles is strongly dependent on the molecular concentration and the thermal-hydraulic conditions. The average size can vary between 0.01 micrometers and 20 micrometers or more. Very small particles are, however, subject to strong agglomeration due to their high number concentration (number of particles per unit volume), and they will rapidly grow to an average size of about 0.5 micro-meters. For typical con-centrations in a reactor coolant system and even in a

containment, the particles will continue to grow but with a slower pace if the residence time is longer than a few minutes. The growth is mainly limited by the decrease in concentration, either by leakage or by deposition.

However, strong formation of aerosols can occur also in the containment if the reactor tank has been penetrated by the molten core. During interaction between the core debris and the concrete floor the temperature becomes high due to chemical reactions. The generated aerosols have generally a larger average size, and they are mainly non-radioactive, but some of the remaining fission products will also be released during this core-concrete interaction. The non-radioactive aerosols will generally increase the deposition rate of all aerosols since they agglomerate by collisions.

The aerosols are transported to other regions in the primary circuit or to the containment due to gas streaming which also includes strong circulational currents. If the velocities are high, deposition is enhanced by impaction, especially in bends and on rough surfaces. Normally, the strongest cause for deposition is the gravitational force, which causes settling on horisontal surfaces. Deposited particles stick to each other and to the surface by molecular forces. If resuspension occurs, the average particle size will then be larger than for the original particles and the consequences may be mitigated by a high deposition rate.

Aerosols can also be removed by water sprays and by driving the aerosols through a water pool, for example the suppression pool in a boiling water reactor. These so-called engineered safety features are the most efficient ones when they are working. However, the natural processes are often sufficient to decrease the release rate to the environment by orders of magnitude. An important factor for achieving such a high degree of decontamination is the time during which the containment is intact. For most of the accident scenarios generally taken into account this time is 24 hours or more. For residence times of the order of a few hours or less, sufficient removal of aerosols can generally only be achieved by engineered safety features. The Swedish and Finnish reactors are equipped with filters which have not been considered in this study.

Aerosol physics is an established science which, however, presupposes a known size distribution of the particles generated and known and stable thermal-hydraulic conditions. Theoretical models have been developed for all known processes, the most important of which are described in the report.

The most complicated equations are concerned with agg-

lomeration and only numerical solutions are generally available. In the 1960's and the beginning of the 1970's the computers were still so slow that a very approximate solution method was developed in order to shorten the computing time, giving rize to the so-called moment-generating codes. Two such codes have been investigated in the AKTI-160 work, namely HAARM-S and RETAIN-S. The more avanced of those codes is RETAIN-S. In contrast to HAARM-S it is for example capable to handle an arbitrary streaming between a large number of compartments and condensation and vaporization both internally in the atmosphere and between the walls and the atmosphere.

In the Nordic project, the results from moment-generating codes have been compared to results from a more accurate solution using the NAUA code. This code is developed by KfK in Karlsruhe and solves the aerosol transport equation by dividing the particle size distribution into a number of classes. A model for the hygroscopicity effect has been developed in Finland in co-operation with the Electric Power Research Institute in USA and has been implemented in NAUA with the version named NAUA-HYGROS. The result of the comparisons was that the moment-generating codes generate a too high value for the aerosol peak concentration and too low values when the concentration has decreased significantly by deposition. This result is valid for dry aerosols. Only NAUA was capable to handle water condensation on particles. The hygroscopicity effect is very important when the relative humidity is close to one or higher.

The above-mentioned codes use thermal-hydraulic data given in the input. In reality there is a strong interaction between the fission product transport and the thermal-hydraulic conditions. This is considered in the MAAP code which handles all known important phenomena during a severe accident. In order to save computing time the particle size distribution is not considered explicitly in MAAP. Instead the code applies empirical correlations for quantities such as suspended mass concentration. The correlations are derived from experiments and from model calculations with more detailed codes. Within the AKTI-160 work results from MAAP- and NAUA-calculations have been compared. It was concluded that the accuracy with MAAP is sufficient for dry aerosols, but the results are inferior if water condensation can occur.

A number of internationally organized experiments have been performed in order to test the aerosol codes for conditions which are close to those of a severe accident. One such experimental series was the Marviken Aerosol Transport Tests (ATT) which was concerned with the behaviour in the primary circuit. The temperature and the aerosol concentration had much higher values than in

any other tests performed. Two experimental series' concerned with the behaviour in the containment and in pipes were DEMONA at Frankfurt am Main and LACE at the Westinghouse Hanford Laboratory in Washington State. Comparisons between experimental and calculational results reveal that the agreement between experiments and calculations was generally poor for the Marviken ATT. The behaviour in the containment as obtained in DEMONA and some of the LACE tests were reasonably well predicted, but the hygroscopicity effect was sometimes of decisive importance. This effect can only be modelled in a few codes, one of which is NAUA-HYGROS. Further, the aerosol behaviour in pipes was generally poorly predicted, but a new model which was firstly introduced in TRAP-MELT, developed at the Battelle Columbus Laboratories, represents a substantial improvement.

Pool scrubbing has also been touched upon in the AKTI-160 work. Two codes which model this mechanism are known, namely SPARC from the Battelle North West Laboratories and SUPRA from the US Electric Power Research Institute. Experiments have been performed at the Battelle Columbus Laboratories, and comparisons between calculations and experiment have been made. It was concluded that the SUPRA model is more complete and gives better results when the gas humidity is high, but both codes underestimate the pool scrubbing effect.

The work has also included an assessment of the present status of knowledge. Much of the information summarized above has been used for pointing out the areas where the models have to be improved, but it is also concluded that perhaps the largest source of uncertainty in the aerosol leakage to the environment is the input data base for the aerosol codes. Also, the consideration of the coupling between thermal-hydraulics and aerosol physics is not satisfactory if the external source term should be calculated with an accuracy of one order of magnitude and the release to the environment is low. Fortunately, the results are generally conservative, that is, the depositions obtained under experimental conditions have been higher than those predicted. During a hypothetical reactor core melt accident the conditions are much more complicated and the conclusions from comparing computations with experiments may not always be valid.

SAMMANFATTNING

Vid en svår reaktorolycka där härden smälter produceras aerosoler, dvs. luftburna partiklar, i riklig mängd redan då härden är överhettad. Dessa består då till stor del av fissionsprodukter och annat material från bränslet. Läckaget av radioaktiva ämnen till omgivningen är till största delen bestämt av aerosolernas beteende. En del aerosoler följer med gasströmmen medan andra deponeras på ytor i primärkretsen, dvs. reaktortanken med kylsystem och ånggeneratorer. Deponerade aerosoler som innehåller fissionsprodukter åstadkommer en höjning av yttemperaturen och kan därför i vissa fall leda till att de återförångas. En stor del av aerosolerna hamnar emellertid i reaktorns inneslutning och kan deponeras där. Detta kan ske både före och efter en eventuell genomsmältning av reaktortanken, som uppkommer om tillräcklig mängd av en smält härd hamnar på tankbottnen. Vid tankgenomsmältning kan härdsmältan i vissa fall hamna på ett betonggolv och åstadkomma en betong-smälta reaktion som ger upphov till ytterligare aerosoler, dels av betongmaterial, men också av fissionsprodukter och annat härdmaterial.

Partiklarna har en storleksfördelning, som beror dels av materialet, dels av de termohydrauliska förhållandena. I primärkretsen kan deras medeldiameter variera mellan 0.001 och 20 mikrometer eller mer. Aerosoler från en betong-smälta reaktion har i allmänhet en större medeldiameter. Om medeldiametern är mycket liten sker en snabb agglomeration av partiklarna på grund av det stora antalet partiklar per volymsenhet. Agglomerationen fortsätter i en långsammare takt om partiklarnas uppehållstid i ett givet utrymme överstiger några få minuter. Tillväxten bestäms av aerosolkoncentrationen, som i sin tur är begränsad av läckage och deponering på ytor.

Den viktigaste orsaken till ytdeponering är normalt gravitationens inverkan. Därför deponerar stora partiklar snabbare än små och ett tillskott av större partiklar från icke-radioaktiva material, t.ex. betong har en gynnsam effekt. Likaså ökas deponeringshastigheten starkt om vatten kan kondensera på partiklarna. En annan orsak till deponering är diffusion, som beror på slumpvisa riktningsförändringar i partiklarnas rörelser. En tredje orsak är partikelmassans motstånd mot riktningsförändringar i rörelsen, s.k. tröghetseffekter. Dessa är verksamma vid snabb olineär strömning (t.ex. i rörkrökar).

För att beräkna aerosoltransporten användes teoretiska eller empiriska modeller. Härvid beskrives ovannämnda processer samt t.ex. agglomeration och läckning med hjälp av matematiska formler, som programmeras i datorkoder. Normalt antages att processerna är oberoende av varandra,

samt att aerosolkällan och de termohydrauliska villkoren är kända. Om källan inte är känd kan den beräknas utgående från modeller för homogen och heterogen nukleation. Vid homogen nukleation formas kondenserade agglomerat av molekyler av ett givet ämne. Vid heterogen nukleation kondenserar ett förångat material på aerosoler av ett annat material. Processerna är starkt beroende av de termo-hydrauliska förhållandena och vid beräkningarna måste denna växelverkan beaktas.

De ekvationer som beskriver aerosoltransporten måste normalt lösas med hjälp av datorprogram. På grund av deras komplicerade natur har man utvecklat olika approximationer. De flesta program använder mekanistiska modeller där sådana finns att tillgå, dvs. modeller som utgår från grundläggande fysikaliska lagar. Men beräkningstiderna blir då långa om tillräcklig noggrannhet skall kunna uppnås. Ett undantag är MAAP-koden, som utvecklats genom samverkan mellan kraftbolag i USA och Norden. Denna kod beskriver alla processer som bedömts vara viktiga vid ett svårt reaktorhaveri, och beaktar härvid i viss mån växelverkan mellan aerosoltransport och termo-hydraulik. I MAAP har aerosoltransporten beskrivits med dimensionslösa korrelationsekvationer, dvs. ekvationer som grundar sig dels på teoretiska modeller med beaktande av resultat från mekanistiska koder, dels på experimentella resultat och som är till stor del oberoende av variablernas storlek. En viktig del av arbetet inom AKTI-160 har bestått i jämförelse av resultat från olika koder, med speciell hänsyn till MAAP-resultat. De noggrannaste resultaten har härvid bedömts komma från koden NAUA, som placerar partiklarna i ett stort antal klasser, som beror på deras storlek. NAUA har utvecklats vid Kernforschungszentrum Karlsruhe i samband med ett omfattande experimentellt projekt. Vid samarbete mellan Finland och Electric Power Research Institute i USA har en förbättrad version av NAUA framtagits, som beaktar kondensation av vatten på hygroskopiska partiklar (NAUA-HYGROS). Motsvarande modell i MAAP har visat sig vara otillfredsställande.

För utvärdering av de teoretiska modellerna har ett antal internationellt organiserade experiment utförts. Vid Marviken Aerosol Transport Tests (ATT) har man simulerat förhållandena i primärkretsen vid ett svårt reaktorhaveri. Vid DEMONA- experimenten i Frankfurt am Main och vid LACE vid Hanford-laboratorierna i Washington State har aerosoldepositionen i en inneslutning undersökts. Vid LACE har tester också gjorts på aerosoltransporten i rör. Internationella jämförelser har gjorts mellan experimentella och beräknade resultat. Inom AKTI-160 har för jämförelse använts koderna NAUA, MAAP, HAARM-S och RETAIN-S. De två senare koderna bygger på en matematisk approximation vid lösning av aerosoltransportekvationen och har ursprungligen utvecklats i USA, men förbättrats i

Sverige. Ett generellt resultat av beräkningar med alla de olika koderna är, att aerosolkoncentrationen minskar med åtminstone tre till fyra tiopotenser om uppehållstiden i en inneslutning är ett dygn eller mer. Detta leder naturligtvis till motsvarande minskning i utsläppet jämfört med fritt läckage till omgivningen av alla de aerosoler som genererats.

Studier har också gjorts på nukleationsprocesser samt på modeller för tvättning av en gas med aerosoler i en vattenbassäng. Experiment för undersökning av tvättningseffekten har gjorts vid Battelle-Columbus Laboratorierna och för jämförelse med dessa experiment har utvecklats koden SPARC vid Battelle North-West i Hanford och koden SUPRA vid EPRI. Beräkningar har gjorts inom projektet med båda dessa koder. Det har visat sig att båda underskattar tvättningseffekten. I MAAP används data för tvättningseffekten i en kondensationsbassäng, som framtagits genom beräkningar med SUPRA. Det är emellertid ej klart om tillämpningen gjorts på rätt sätt i de nordiska versionerna av MAAP.

Ett försök att uppskatta felgränserna vid beräkningar av aerosoltransporten har också gjorts. Tillräcklig noggrannhet har uppskattats vara en faktor tio om den genererade aerosolmängden är 10 000 gånger större än den som läcker ut till omgivningen. På grund av stora osäkerheter i in-data (termo-hydrauliska förhållanden) kan denna noggrannhet knappast uppnås, men deponeringen torde i allmänhet underskattas vid beräkningarna. En viktig faktor i detta sammanhang är också växelverkan mellan aerosoltransport och termo-hydraulik. Inom detta område bör ytterligare utvecklingsarbete ske.

De viktigaste slutsatserna, som framkommit inom AKTI-160 är följande:

Den bästa överensstämmelsen mellan beräkningar och experiment har erhållits med koden NAUA-HYGROS. MAAP ger i allmänhet tillfredsställande överensstämmelse för torra aerosoler. För båda koderna var resultaten vid beräkningar på torra aerosoler konservativa. MAAP kan ge för hög deponering under förhållanden med hög fuktighetsgrad. HAARM-S och RETAIN-S ger sämre överensstämmelse med experiment.

Förhållandena vid en verklig reaktorolycka är så komplicerade, att inga beräkningar med säkerhet kan ge tillräcklig noggrannhet för mängden aerosoler som läcker ut till omgivningen. Gruppens arbete har ej omfattat inverkan av filter, som ju är avsedda att kompensera för den nämnda osäkerheten.

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1. INTRODUCTION

In the plan for NKA's nuclear safety program 1985-1989 (NU 1984:13), the prospect for AKTI-160 was to develop a fast calculational method for aerosol transport which would be more accurate than that used in e.g. RETAIN, or other codes with a log-normal distribution for the particle sizes. It was, however, realized at an early stage that such a project would be too expensive in view of the limited resources available (LAU85a). Instead, another approach was proposed:

- Examine the possibilities of running NAUA with fewer size groups, thus saving computer time without losing too much accuracy of the results.
- Examine other possibilities for speeding up NAUA, e.g. improving the integration technique.
- Extend the code comparison exercise slightly by including a few runs with the aerosol part of the MAAP code in order to assess the validity of using correlations.
- Study the importance of mechanisms which are not at present considered by all of the codes.

The main course of this proposal has been held. Thus, comparative calculations have been performed between codes using the assumption of log-normal particle size distribution and codes with discrete size distribution, and calculations have also been made with a module of MAAP-3 which solves the transport problem by a correlational method. Further, the group has participated in an international comparison of calculations on the DEMONA experiment. Studies have also been done on the nucleation process, on hygroscopicity, and on pool scrubbing. Nucleation is implemented in the RAFT code as an important mechanism. Simulations with this code in the Marviken-V experiment indicated its practical importance. The hygroscopicity of CsOH has a strong influence on the experimental results in LACE. It has been implemented in NAUA by J. Jokiniemi. Pool scrubbing is one of the most important decontamination mechanisms inside the containment in a boiling water reactor.

Finally, the international experimental and theoretical research has been followed up by reporting from conferences and by literature searches. It has been concluded that the codes with discrete particle size distribution have been established as a standard. New and/or improved models have been introduced in several codes. The importance of chemical processes has been recognized. A new code which may predominate in the future is VICTORIA, developed at the Sandia Laboratories for the US-NRC. Important mechanisms in this code are heating by fission products, chemical reactions and inter-volume gravitational settling. According to D.A. Williams, AEEW, the latter mechanism replaces nucleation as an

explanation for the large particles found in the Marviken-V experiment (private communication). The opinion of the people who developed the RAFT code is, however, different and this is still a point of discussion. Further, the models for turbulent deposition and deposition in bends have been improved. A phenomenon which remains relatively unexplored is resuspension.

After some introductory paragraphs, the report describes the phenomenology in aerosol behaviour. Then the international background is given. Calculations for intercomparison between codes and for comparison with DEMONA are described. The work dealing with nucleation, hygroscopicity and pool scrubbing is reported in paragraphs 5.2, 5.3 and 5.4, respectively. Finally, an assessment of the present status of knowledge in this field is made in section 6. This is based partly upon the calculations and partly on the literature search.

2. OBJECTIVES AND ORGANISATION OF THE PROJECT WORK

The AKTI-160 project group was formed early in 1985. The commission was to evaluate the aerosol transport codes used in the Nordic countries.

The project group has been free to choose its tasks. It soon turned out, however, that there was an overlap with the project groups AKTI-130 (Sensitivity studies) and AKTI-150 (Chemistry) which required some coordination and collaboration. The project group has met about twice a year and the findings have been described in a number of technical reports and a few publications. The group members have also taken part in other work on nuclear aerosol behaviour.

The evaluation has taken place in a number of ways. Selected parts of the codes have been scrutinised, the codes have been compared to each other in benchmarking exercises and they have been compared to experiments.

The codes considered include one or more versions of HAARM, RETAIN, MAAP, TRAP-MELT, RAFT and NAUA. Some of these codes have also been modified either for corrections or for applications to specific purposes. Most of the codes are available through the NEA Data Bank or through the IAEA, but MAAP, which is used extensively in Sweden and Finland and RAFT are proprietary.

The report reflects the project status in the Spring of 1989 and was written as a joint effort by the following members of the group:

Peter Fynbo	RISOE NATIONAL LABORATORY, Roskilde, Denmark
Hans Häggblom	STUDSVIK NUCLEAR, Nyköping, Sweden
Jorma Jokiniemi	VTT, Helsinki, Finland

The late Kaj Erik Lindström Jensen, ELSAM, was a member of the group in 1985. Kurt Lauridsen was a member of the group until Jan. 1, 1988, but moved at that date to other tasks at the Risoe National Laboratory.

Jouni Mäkynen and Aimo Hautojärvi have been deputees during the Jorma Jokiniemi's leave of absence.

Credit for the work and responsibility for the opinions expressed rests with the above mentioned authors.

3. PHENOMENOLOGY OF AEROSOL BEHAVIOUR

An aerosol is a stable or quasi-stable system of liquid or solid particles suspended in a gas. Aerosol physics is important in LWR accident analysis, because the main part of the fission products released from the core during an accident will form an aerosol by condensation or mechanically from the bubbling melt. The largest part of the aerosol material will only be slightly radioactive or non-radioactive, as it stems from structural materials, unused fuel or - in the containment - from concrete or steam. Noble gases do not form aerosols and are less important biologically, but the gas phase may contain a small fraction of the iodine. However, noble gases and other gas phase fission products in the containment atmosphere will not be considered in the following.

During a reactor accident a number of processes may produce aerosols:

The gap release results from fission product vapours in the pellet-cladding gap, in pellet-pellet interfaces, and in pellet cracks. At temperatures of 800°C - 1100°C (that is, before melt) cladding may fail owing to internal overpressure so that volatile fission products (including noble gases) will escape. This is called "burst release". Also included in gap release is a slower release by diffusion in the gas phase through interconnected voids in the fuel. The gap release comprises at most a few per cent of the volatile fission products.

Diffusion release from the solid fuel is important at temperatures above 1400°C. The main processes are diffusion in bulk material and in grain boundaries. Porosity is important and itself dependent on the dynamics of the bubbles of fission product gases.

Melt release occurs at temperatures above the melting point of the fuel. It takes place by diffusion in the molten material and in the boundary layer. The vapour pressure of the radionuclide in question is assumed to be the decisive parameter.

It should be mentioned at this point that the melting point of the fuel depends on the accident sequence. For example, the fuel may interact with cladding to form a Zr-UO_2 melt. The degree of this interaction depends on parameters such as temperature vs. time and the amount of available steam.

Sparging (also known as vapourisation) occurs during the corium-concrete interaction (CCI). Gases are produced by decomposition of the concrete and particles are formed mechanically as the gases sparge through the melt, and by vapourisation. Most of the aerosol material produced by sparging will be non-radioactive.

Other mechanisms are oxidation in the case of steam explosions, and revapourisation. Revapourisation is caused by the decay heat developed by deposited radionuclides and will begin at about 700°C. It may be important for radionuclides deposited on poorly cooled surfaces in the primary system. The importance, however, depends on the possibility of transport. If there are two holes in the primary system a chimney effect may be important. If revapourisation takes place when containment fails, fission product vapours will flow from the primary system into containment. However, it is also possible that revapourisation will simply result in recondensation on "cold" surfaces in the primary system.

Very little has been published on the size distribution of the particles produced by the above-mentioned processes. In experiments (ALB84) the median diameter of particles from core melt in steam was found to be 0.09 μm and the geometric standard deviation was 2.13 (log-normal distribution). The particles produced by CCI are larger; a mean aerodynamic diameter of 2 μm and a geometric standard deviation of 2 are given by Wichner et al. (WIC81) for a log-normal distribution.

The origin of aerosols and an example of a size distribution function is shown in fig. 3.1.1. Fig. 3.1.2 shows the phenomena contributing to fission product transport.

The aerosol of a reactor accident may be very dense. In the containment the peak density may be higher than 20 g/m^3 and in the reactor pressure vessel the peak density may be of the order of 100 g/m^3.

Dense aerosols decay fast. This can be understood, when it is recognised that the agglomeration rate is quadratic in the particle number density. Loosely speaking: the more particles initially, the bigger particles later on. When it is further recognised that settling is usually the dominating removal mechanism and that bigger particles settle faster than smaller ones, other things being equal, it is seen why dense aerosols decay fast. A typical case for a dry aerosol in a PWR containment shows depletion by three orders of magnitude during one day and a further three orders of magnitude during the next five days (HAS87).

Another important characteristic of LWR containment aerosols is the presence of saturated or near-saturated steam. This has a number of consequences:

- owing to surface tension, vapour pressure at a particle surface depends on the particle radius (Kelvin

effect). Therefore, particles below a critical size are dry and do not grow. Because of this, a size distribution curve will have two "humps" in a supersaturated atmosphere.

- particles are nearly spherical, possibly because of surface tension. Therefore, form factors can be set to 1 in calculations, reducing the uncertainty.

- particles become heavier owing to steam condensation. This generally enhances removal by settling.

- the steam condensing on walls gives rise to an additional removal mechanism, diffusiophoresis.

Only the amount of fission products suspended in the containment atmosphere is available for an activity release to the environment. It is therefore important to obtain estimates of the retention capacities of the primary system and of the containment. The retention results from a number of "removal mechanisms", which transport the aerosol particles to some surface where they stick, usually as a result of van der Waals forces.

The physical processes that operate on an aerosol are many. The more important ones for LWR safety are:

a. Gravitation

In most of the cases considered, gravitational settling (=sedimentation) is the main removal mechanism. Larger particles fall faster than small particles and thus are removed sooner. Also, they can sweep up smaller particles when falling. This is called "gravitational agglomeration".

The importance of sedimentation can be illustrated with the results from the DEMONA benchmark exercise referred to in section 4.3 below. In this exercise sedimentation is responsible for the deposition of about 10 kg, diffusiophoresis is responsible for about 2 kg and diffusion for only about 10 g. The ratios between these numbers may vary according to circumstances, but the result gives an indication of the relative importance of the removal mechanisms.

b. Brownian diffusion

This is important for small particles. It causes deposition on walls, floor and ceiling. It also contributes to agglomeration.

c. Diffusiophoresis

Aerosol particles are pushed towards walls where steam condenses, owing to the net gas flow in that direction. (Strictly, "Stefan flow" is the correct term for this process).

d. Thermophoresis

Aerosol particles in a gas temperature gradient will move towards the lower temperature.

e. Effects of carrier gas flow and particle inertia

The obvious effect of gas flow is transport from one compartment to another. However, flow may influence an aerosol in other ways. Particle removal will be increased because the flow brings fresh particles to volumes where the aerosol is depleted. If the flow is laminar and changes direction, inertia also increases the removal. Further, particles will deposit in turbulent flow because of the fluctuating velocity components normal to the surfaces.

In laminar flow fast-moving particles will catch up with slower particles on neighbouring streamlines and thus cause "shear agglomeration". Shear agglomeration is also important in turbulent flow where it is further augmented by particle inertia.

The gas flow can also cause resuspension of deposited particles. This phenomenon is not sufficiently investigated but it is believed to have only small importance.

f. Homogeneous nucleation

If the saturation ratio is "large", vapour or steam may form a large number of small, new particles. Owing to surface tension, particles below a certain size are not stable (Kelvin effect). The critical size depends on material, temperatures etc.

g. Condensation

Steam and fission products can condense on or evaporate from the particles. The condensation (evaporation) rate depends on the saturation ratio which in turn is determined by condensation on (evaporation from) both particles and structures. Obviously, condensation and evaporation depend strongly on the thermal conditions. Certain materials are hygroscopic and require methods

from chemistry for a proper treatment. For our purposes the most important example is CsOH which will be considered in section 5.3 below.

To limit the buildup of containment pressure, nuclear power plants are equipped with one or more emergency safety features (ESFs) in the containment.

Containment ESFs may be passive, such as the pressure suppression pools applied in BWRs, or they may be active, such as containment sprays. Ice condensers may be active or passive. A new class of ESFs is the filtered containment venting systems (FCVs) installed or to be installed in a number of nuclear power plants. FCVs can be active or passive. All these ESFs have, beside their main purpose of reducing containment pressure, the effect of reducing the source term. In pressure suppression pools aerosol particles are caught by the bubble surface. The mechanisms are diffusion for small particles, impaction for large particles and diffusiophoresis. Most of the above-mentioned removal mechanisms come into play for both ice condensers and containment sprays. For an FCV the source term reduction is evidently essential, irrespective of the design.

The relative contributions from the different deposition mechanisms depend on the sizes and shapes of the particles, the gas velocity and composition, and the geometry. An illustration was given in a famous report to the American Physical Society (PIN85) where various deposition velocities as function of particle diameter were calculated for aerosol transport in a pipe. The results are shown in fig. 3.1.3. The turbulent deposition corresponds to a gas flow with Reynolds number of 2×10^5.

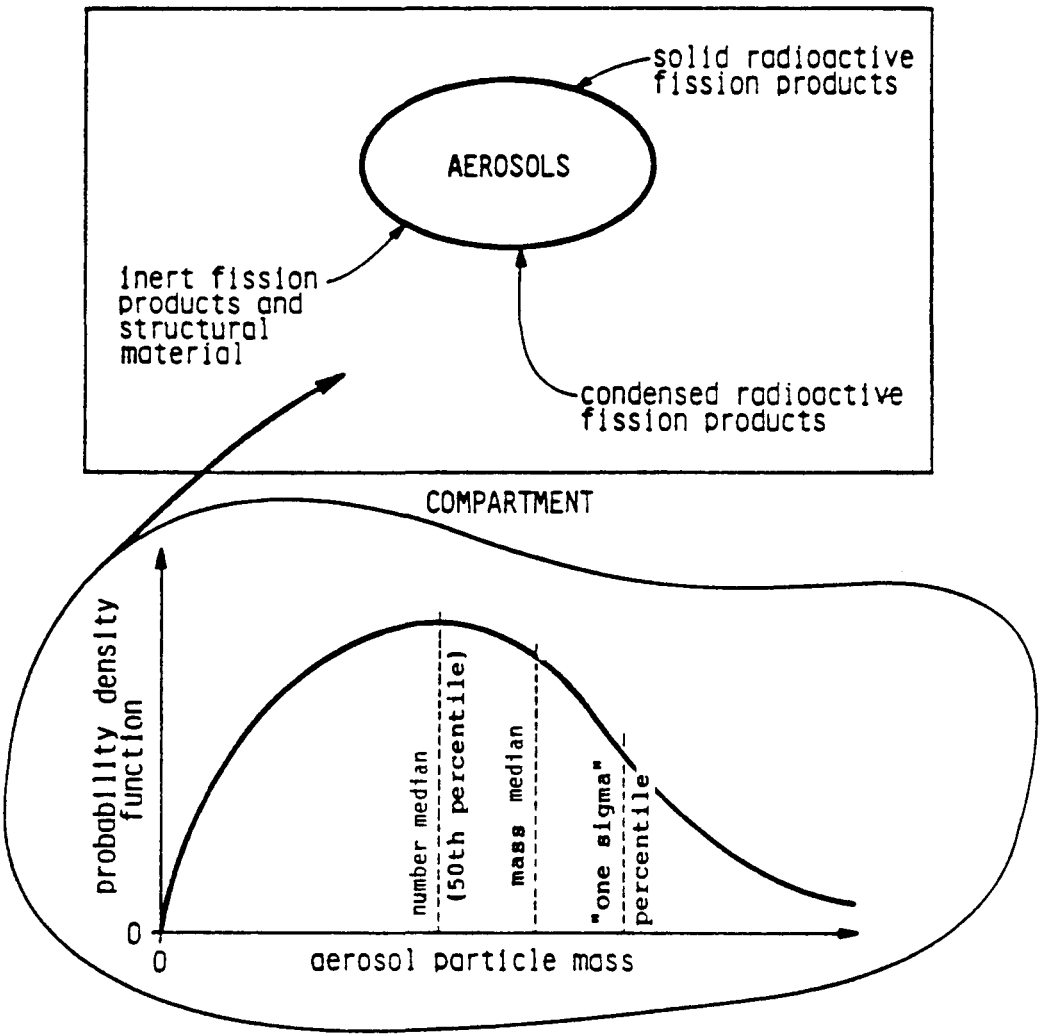


Fig. 3.1.1 The origin of aerosols and an example of a size distribution function.

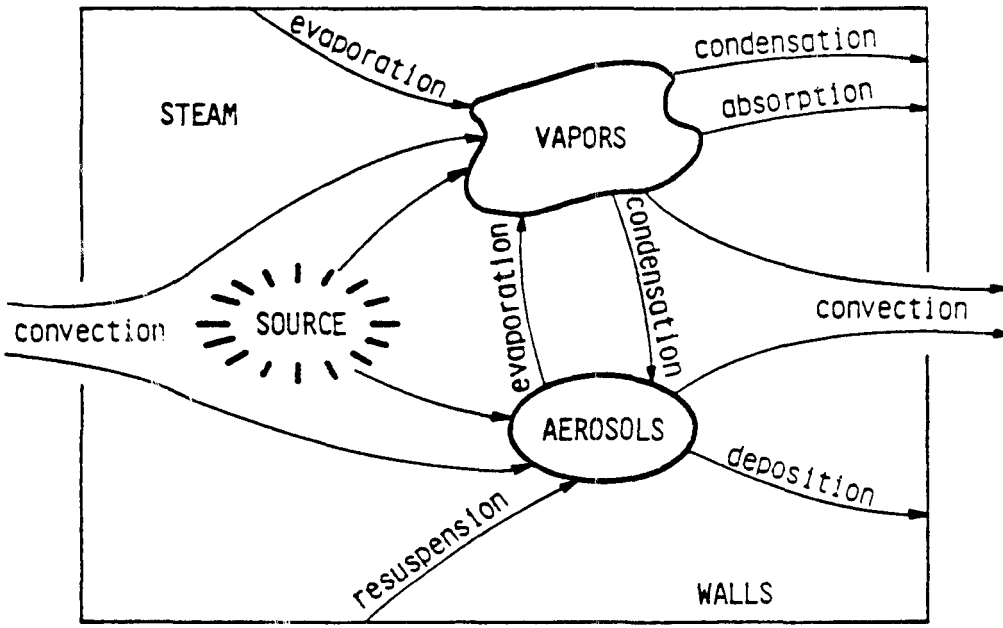


Fig. 3.1.2 The phenomena contributing to fission product transport.

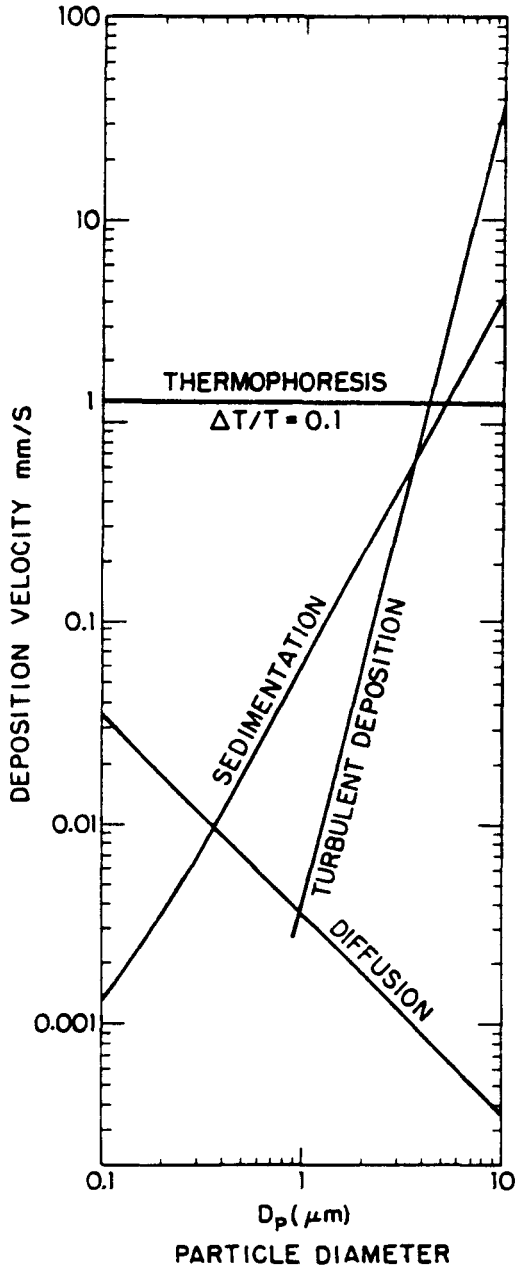


Fig. 3.1.3 Various deposition velocities as a function of particle diameter, D_p, for a hypothetical forced convection example.

4. THE INTERNATIONAL BACKGROUND

4.1 Overview

For the classical US Reactor Safety Study, WASH-1400, the aerosol behaviour was not considered in a generic, mechanistic way. A full scale study, CSE, was however performed at the Pacific North West Laboratories (HIL70). The results were used for developing a computer code, CORRAL, with correlational models for the fission product deposition in a containment. Experiments for a more fundamental understanding of the aerosol behaviour were initially concerned with LMFBRs. Research on aerosol transport under conditions typical for an LWR containment were started at Karlsruhe under the NAUA project (HAU76). As a demonstration of the validity of the results from the NAUA project, the German-Swiss DEMONA experiments were carried out (HAS89). After the TMI-II accident large scale research started in the USA where EPRI initiated both the Marviken-V project (MAR85) and the LACE experiment (WIL87), although they were internationally funded. The US-NRC was a co-sponsor to these projects and has also sponsored small-scale aerosol experiments at national laboratories.

Based on theory and on the experiments different aerosol computer codes have been developed. The most well-known of the established codes are:

Name	Developed by
AEROSIM	UKAEA, UK
AEROSOLS/B1	CEA, France
CONTAIN	Sandia Lab., USA
NAUA	KfK, FRG
PATRAP	JAERI, Japan
TRAP-MELT	BMI, Columbus, USA

MAAP is an integrated thermal-hydraulic and fission product transport code with a correlational model for aerosol deposition. It is owned by EPRI and has restricted availability. Codes with approximate solutions of the aerosol transport equations are:

HAA-4	Rockwell International, USA
HAARM	BMI, USA
RETAIN	EPRI, USA

More recent codes which are still in the developing stage are:

RAFT	EPRI, USA (developed at ANL)
VICTORIA	Sandia Lab. USA

Many of these codes have been developed further at different laboratories and therefore there are different versions of them.

The most important international work is described briefly below, starting with the experiments. As much of the material is restricted, the intention is only to give a qualitative picture of the work which has been done and the results obtained. For the CSE project no code validation work was done and therefore it is not considered in section 4.3.

4.2 Experiments

4.2.1 The Containment System Experiment, CSE

The information obtained for this experiment is from ref. (HIL70). The program was performed at the BMI Pacific Northwest Laboratories, Richland, during the years 1968 to 1970 and sponsored by the US Atomic Energy Commission. In some of the tests in the CSE, active safety features were operated to study their performance and to verify mathematical models. In other tests all fission product behaviour was due to natural processes. Both iodine and aerosols in the form of cesium and uranium oxide were studied. Initial particle concentrations ranged from 0.1 to 10 mg per m^3 in two different vessels. The vessel volumes were 65 and 750 m^3.

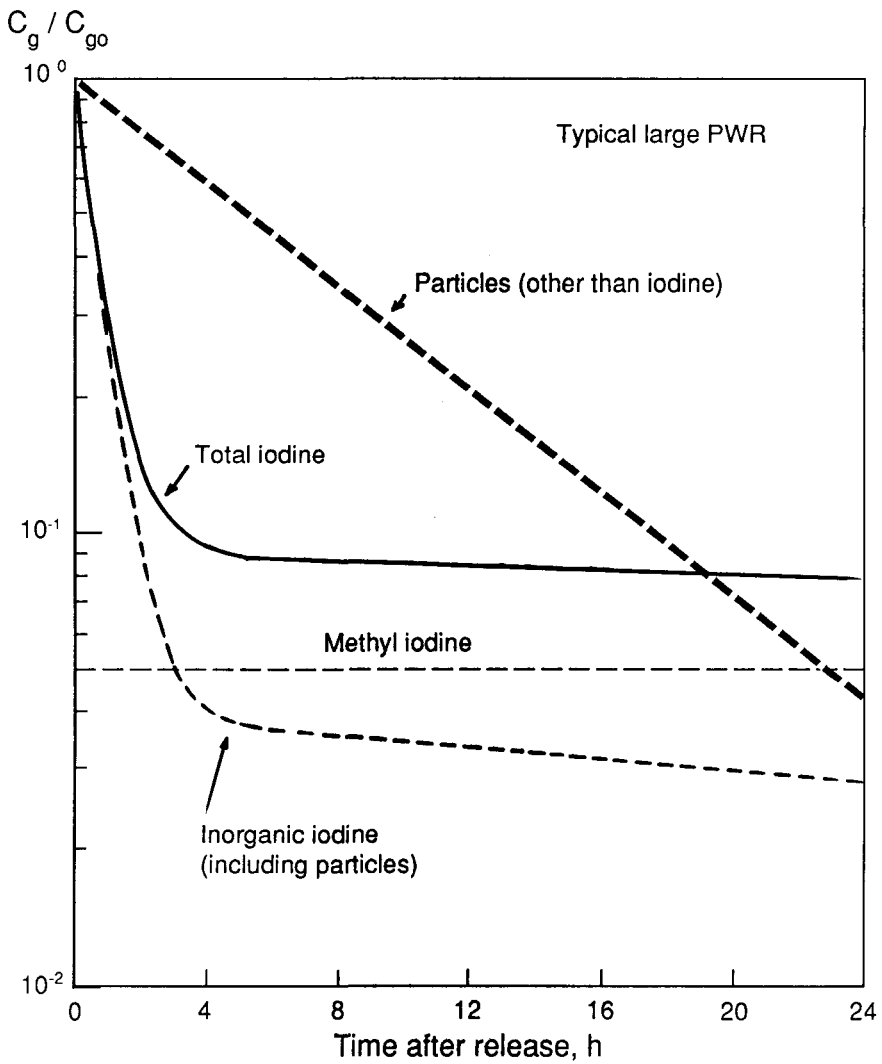
The atmosphere was always supersaturated with steam during the tests. The aerosol deposition rate was measured and used to calculate the particle diameter assuming that the deposition was due to the following mechanisms:

gravitational settling
Brownian diffusion
diffusiophoresis

It was found that the particle diameter was 10 to 15 microns early in the tests, decreasing to 3 to 7 microns after a few hours. It was concluded that the particle size was mainly determined by steam condensation and evaporation. Gravitational settling was responsible for the major portion of the removal rate.

Fig. 4.2.1 shows the predicted decrease in aerosol and iodine concentrations in a typical large PWR as a result of natural removal processes. The experimental results were also used to develop models for both natural removal processes and removal due to engineered safety features. These models were used in the CORRAL code (AEC75).

Fig. 4.2.1 Prediction of Iodine and Particle Mass Concentration in the Containment Atmosphere of a Typical Large PWR



4.2.2 The Marviken Aerosol Transport Tests

The Marviken Aerosol Transport Tests (ATT), (MAR85) are the only experiments done in an LWR full scale primary circuit on aerosol and fission product phenomena during a severe accident. Five experiments were performed: Tests 1, 2a, 2b, 4 and 7. In the first three tests aerosols of CsOH, CsI and Te were injected into a pressurizer followed by a pipe system, a relief tank and a filter. In test 7 the source consisted of the same species but was located in a reactor vessel upstream from the pressurizer. In test 4 this "fissium" source was supplemented by "corium", composed of silver and manganese.

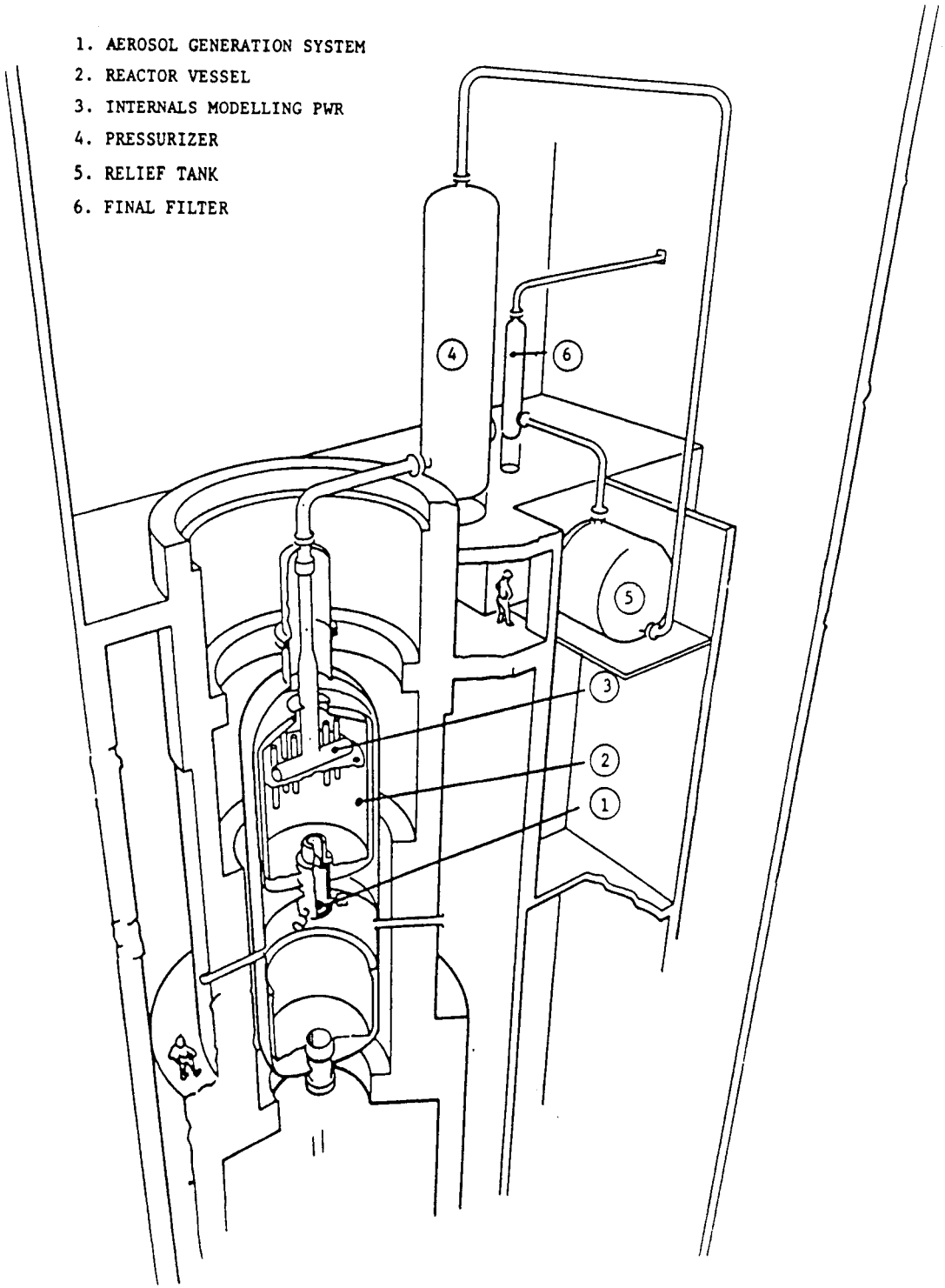
The main characteristics were different from those in other aerosol tests in at least the following respects:
- The temperature in some compartments varied from 300 to 1200°C, as compared to less than 110°C in other tests.
- The aerosol source feed rate was up to 80 g/s, as compared to about 2 g/s in e.g. the LACE tests.
- The residence time in each zone was shorter than in the containment tests.

These characteristics lead to difficult experimental conditions and to more severe demands on the models and computer codes in order to consider all the phenomena of importance. The high temperatures lead to chemical reactions which are considered only in the Japanese codes MPEC and PATRAP, the EPRI code RAFT, and in the recently developed code VICTORIA. The high peak concentration made the nucleation processes important. The short residence time decreased the importance of sedimentation compared to other deposition processes.

Fig. 4.2.2 shows the main arrangement which included:

- Aerosol generation system
- Reactor vessel (volume approximately 137 m^3).
- Reactor internals (surface area approximately 200 m^2).
- Pressurizer (volume approximately 50 m^3).
- Relief tank (volume approximately 50 m^3).
- Piping between principal tanks.
- Final filter.

Fig. 4.2.2



Test facility arrangement

Some experimental conclusions:

- There was a significant deposition in the reactor vessel in spite of the high temperature. Thus, chemisorption was an important deposition mechanism.

- The wall deposition in the pressurizer was about 40% of the total deposition for test 7, otherwise 10-20%. This was in all cases more than predicted.

- Deposition in pipe bends was high.

- In tests 1 and 2 CsOH deposited in the form of a liquid.

- The particle sizes were larger than predicted. For test 4 the size distribution was bimodal and one fraction had an Aerodynamic Mass Median Diameter (AMMD) between 50 and 70 μm. The AMMD is defined as the diameter of a water droplet with the deposition velocity such that the measured deposited mass is obtained.

4.2.3. The DEMONA Research Programme

The DEMONA (DEMOnstration Nuklearen Aerosolverhaltens) research programme served the purpose of demonstrating (not investigating) the behaviour of nuclear aerosols in large scale experiments. The experiments were performed at the model containment of the Battelle-Institut, Frankfurt am Main. The participating organisations were Kernforschungszentrum Karlsruhe, Projekt Nukleare Sicherheit, Kraftwerk Union, Eidgenössische Institut für Reaktorforschung, and Battelle-Institut.

Experiments

The Battelle model containment is a 1:4 model of the Biblis A PWR containment, apart from the dome, which is somewhat smaller than this. Various internal configurations can be chosen ranging from a single compartment geometry of 640 m^3 to a buildup with 9 compartments (see fig. 4.2.3, HOS83, SCH83).

In the experiments the containment was heated to 110-115°C by steam injection. The aerosol was generated by 3 plasma torches of 80 kW each, the generation lasting typically 1 hour.

Of the 9 experiments in the series, 7 were in a condensing atmosphere. The aerosol materials were SnO_2, Ag+MgO, Fe_2O_3, and SnO_2+Fe_2O_3. The peak mass concentrations ranged from 2 g/m^3 in a low-density experiment to 12 g/m^3.

During the experiments both thermal and aerosol-physical parameters were measured, e.g. pressure, temperatures at various positions, leak rate and steam condensation rates at the external walls. Aerosol mass concentration and size distributions were measured by a number of methods, including filters, inertial spectrometers, cascade impactors, and infrared photometers. The water content of the aerosol particles was measured by a calorimetric method.

Results

A convenient single parameter for comparison of experiment and model calculations is the time integral of the aerosol mass concentration, because this integral is proportional to the amount of leaked material.

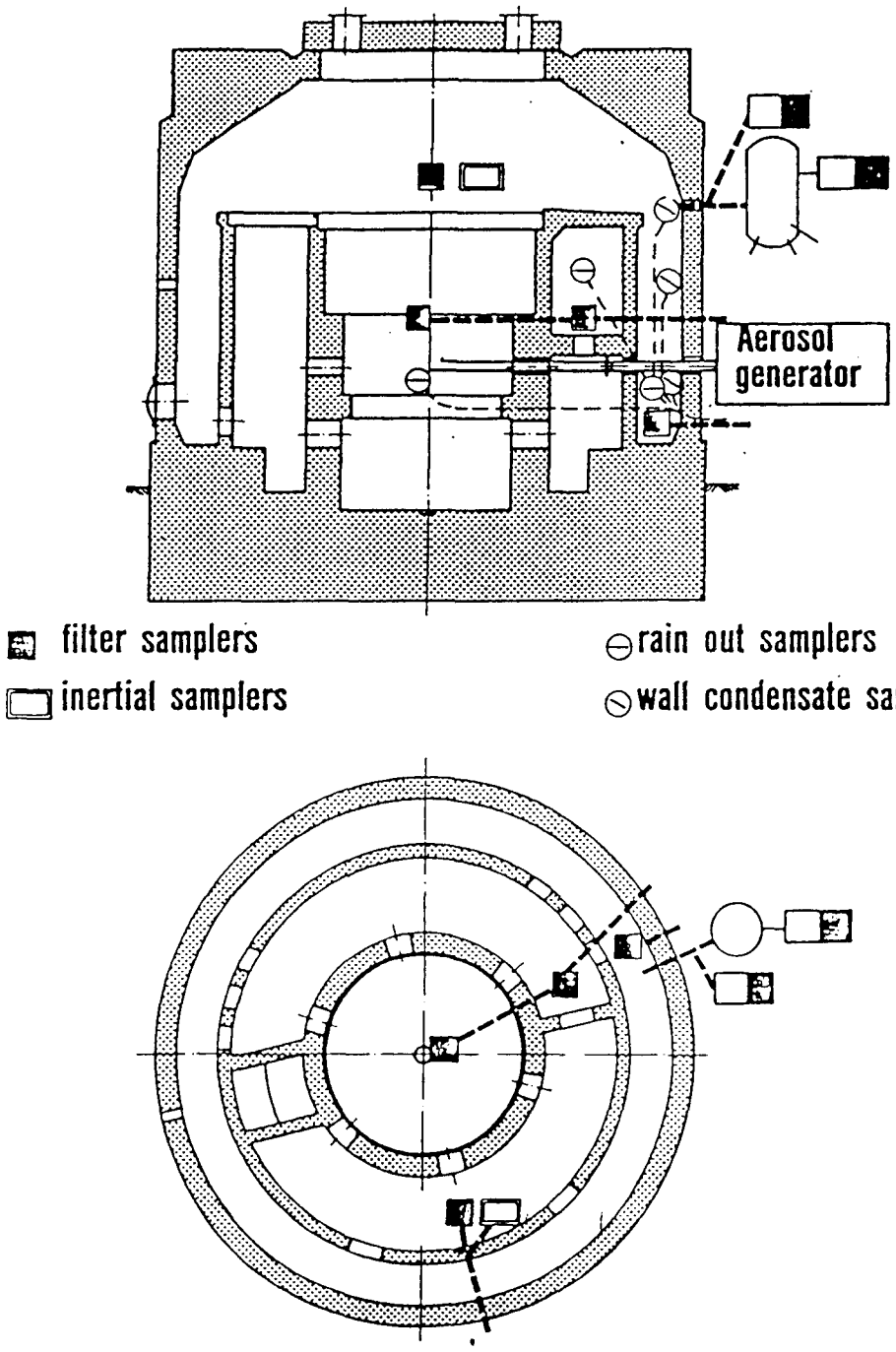


Fig. 4.2.3 The Battelle model containment with positions of sampling stations.

Calculations with the NAUA code consistently overestimate the time integral by factors of 1.7 to 3.0 for the experiments in condensing atmosphere (HAS88). The reason for this is that turbulent and thermal removal mechanisms, which are not modelled in NAUA, are active during the aerosol injection period (SCH88). These mechanisms will not be of the same importance in a full-scale PWR containment, and the conservatism of the code, which might be considered a weakness from the point of view of aerosol physics, can be considered a strength from the point of view of reactor safety, moderate as it is.

It has also been suggested (E. Schrödl, private communication) that the efficiency of the plasma torches is lower than assumed and that this may constitute part of the explanation for the overestimation of the mass con-centration.

4.2.4 LACE

The object of LACE (LWR Aerosol Containment Experiments) was to study aerosol behaviour in pipes and in a containment in conditions expected during severe accidents. LACE was an international project, which was managed by the Project Board (PB) with the support of the Technical Advisory Committee (TAC). The experiments were started in 1985 and finished by the end of 1987. LACE experiments were carried out in the Westinghouse Hanford Laboratory. The experiments and the code comparison work were organised by the Electric Power Research Institute (EPRI).

The LACE experiments can be divided into three parts: 1) pipe experiments, 2) containment experiments, and 3) rapid containment depressurization tests. The experimental arrangements for tests LA1 (pipe and containment experiments) and LA6 (depressurization tests) are described in figs. 4.2.4 and 4.2.5, respectively.

Pipe experiments

Pipe experiments included so-called pretests (CB series), LA1 tests and LA3 tests. In CB tests the aerosol generation, chemical composition of aerosols and thermal hydraulics were tested. The aerosol injected into the pipe was composed of hygroscopic CsOH and non-hygroscopic MnO (in the CB series NaOH and Al_2OH_3). Nitrogen and steam were injected together with the aerosol. The effect of the following parameters on pipe deposition was studied:

a) Gas flow velocity (22 - 145 m/s).

b) Hygroscopic matter mass fraction in the injected aerosol particles (0 - 1).

c) Pipe inlet particle size (1.4 - 4.3 μm)

d) Gas superheat (15 - 183°C)

The resulting deposition in pipes varied between 1% and 98% (Table 4.2.1).

Fig. 4.2.4

EXPERIMENTAL ARRANGEMENT — LACE TEST LA1

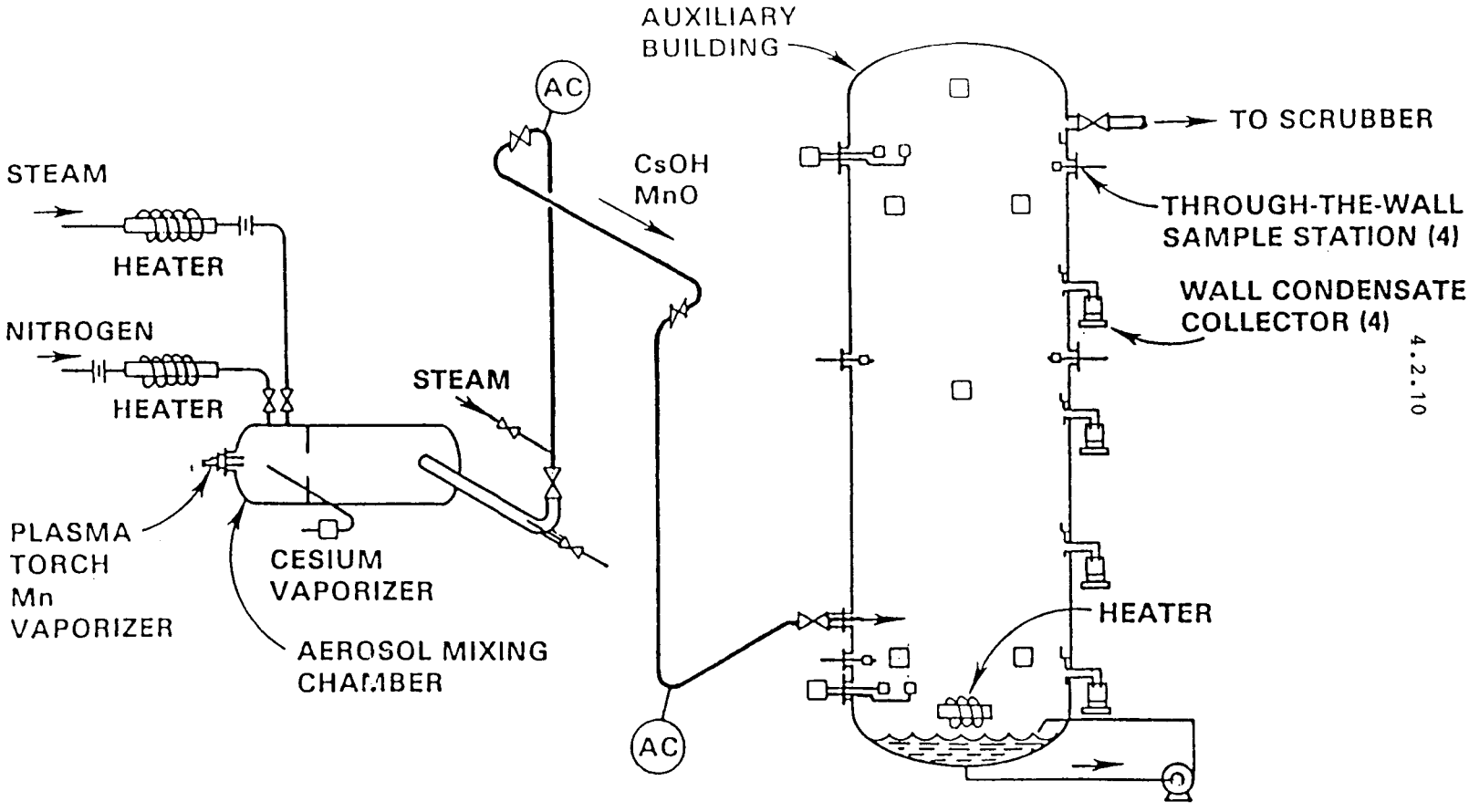
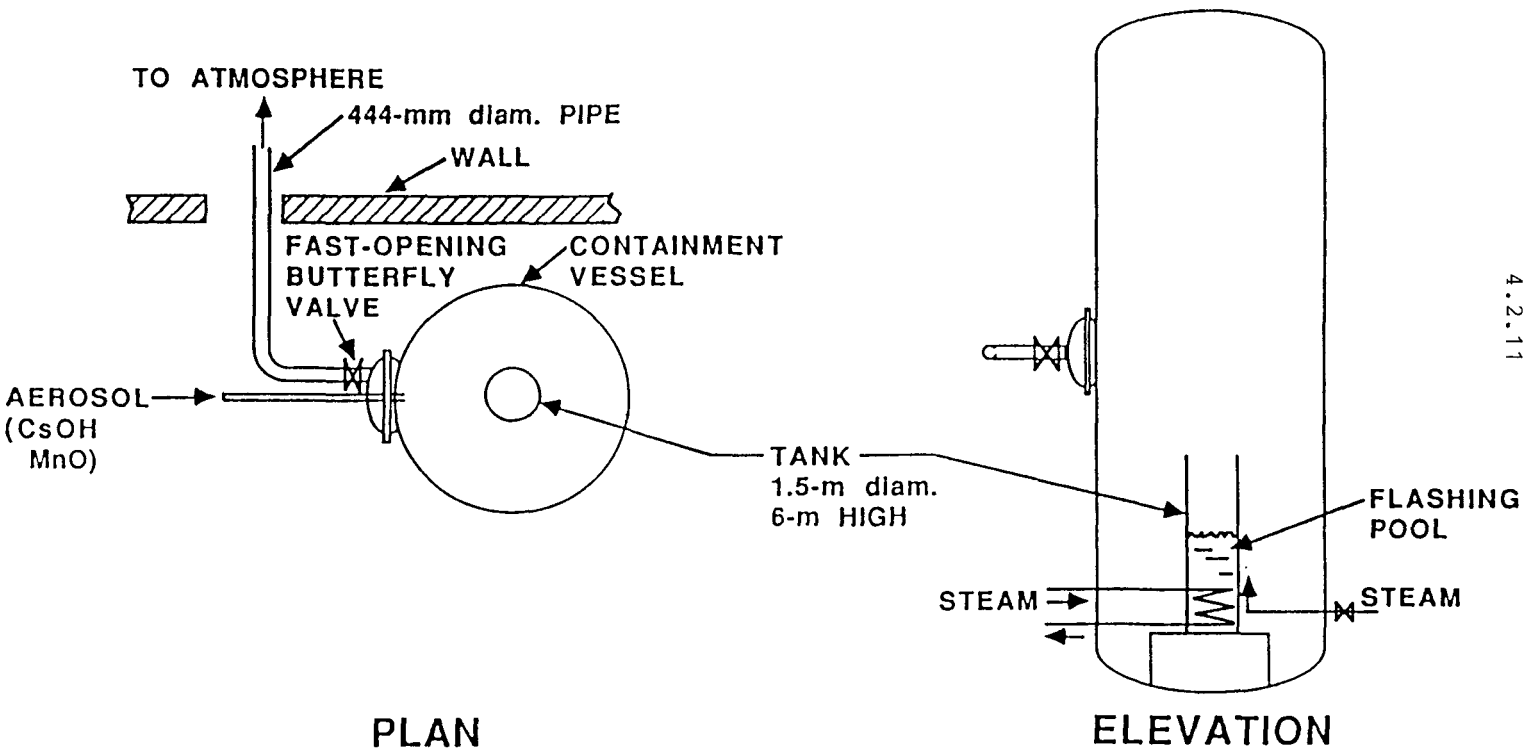


Fig. 4.2.5

TEST ARRANGEMENTS FOR LA5 AND LA6



4.2.11

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SUMMARY OF AEROSOL FLOW THROUGH TEST PIPE

TABLE 4.2.1

	LA1	LA3A	LA3B	LA3C	CB1	CB2	CB3
AVG. GAS VELOCITY, M/S	145	80	23	22	123	106	122
FRACTION HYGROSCOPIC	0.42	0.17	0.12	0.38	1.00	0.59	0
INLET SIZE (AMMD), MICROMETERS	1.7	1.4	2.4	1.9	3.9	3.2	4.3
GAS SUPERHEAT, DEG. C	141	183	151	164	88	15	66
PERCENT OF AEROSOL PENETRATING PIPE	2	23	49	17	97	95	99

Containment Experiments

In the containment experiments (LA1, LA2, LA4 and LA6) the behaviour of hygroscopic (CsOH) and non-hygroscopic (MnO) aerosols in an 852 m^3 vessel was studied. In addition some phenomena could be studied in test LA6 before rapid depressurization (450 minutes). The effect of the following parameters on aerosol behaviour was studied: a) timing of venting b) timing of CsOH and MnO injection c) the amount of injected mass.

During tests LA2 and LA6 CsOH and MnO were injected simultaneously. In the LA4 test CsOH was first injected over a 30 minute period, then CsOH and MnO were injected together for 30 minutes and after that only MnO was injected for 30 minutes. LA1 and LA2 had pre-existing leakages up to 60 and 277 minutes, respectively. In LA4 the vent period was from 280 to 440 minutes. In LA1 the mass entering the vessel was only about 1% of that in the other tests.

Rapid Depressurization

The effect of rapid depressurization on aerosol behaviour, resuspension and dispersion into the vicinity of the test facility was studied in tests LA5 and LA6. In these experiments the pressure was decreased from 3 bars to atmospheric pressure during a period of one minute. In the bottom of the containment vessel there was a water pool (Fig. 4.2.4). This pool also contained water soluble Li_2SO_4 and insoluble ZnO. In addition, in test LA6 aerosol (CsOH and MnO) had been injected into the containment vessel up to 50 minutes. After this the aerosol was aged for 400 minutes in the vessel before depressurization. Only a small part of the resuspended mass was released into the environment. The integrated near field fallout was between 40 - 60 % of the released mass.

4.3 Code validation and benchmarking

4.3.1 The Marviken Aerosol Transport Tests

Within the Marviken project there was no organized activity for code validation and benchmarking, but an unofficial group has been present at the Marviken-V Analysis Specialist's meetings. The laboratories which have contributed with presentations at these meetings are:

- Atomic Energy Establishment, Winfrith (UK)
- Argonne National Laboratory (USA), representing EPRI
- Battelle Columbus Laboratories (USA)
- CEA, Fontenay aux Roses, Cedex (France)
- Central Electricity Generating Board, London (UK)
- Electric Power Research Institute, Palo Alto (USA)
- Ente Nationale per l'Energia Elettrica (Italy)
- STUDSVIK Nuclear (Sweden)
- Tokai Research Establishment, JAERI (Japan)
- VTT, Helsinki (Finland)

The computer codes used by these laboratories for aerosol transport, chemical equilibrium and thermal-hydraulics calculations were:

- AEROSOLS/B1 (CEA) for aerosol transport
- MAAP-3 (STUDSVIK) for aerosol transport
- MPEC (JAERI) for chemical equilibrium
- NAUA-4 (EPRI) for aerosol transport
- PATRAP (JAERI) for aerosol transport
- RAFT (ANL) for chemical equilibrium, nucleation and aerosol transport
- RETAIN-2c (VTT) for aerosol transport
- RETAIN-S (STUDSVIK) for aerosol transport
- SPRITE (AEEW) for thermal-hydraulics
- TRAP-MELT-2 (BCL and ENEL) for aerosol transport
- TRAP-MELT2-UK (AEEW) for aerosol transport

Mechanistic models were used in most codes, with the exception of MAAP-3 which uses correlational models for aerosol transport. Some characteristic features of the aerosol transport codes are given in Table (4.3.1).

Test 1 was performed 1983-10-11. The results diverged greatly between the different codes and also between the different TRAP-MELT calculations. Moreover, the C/E values (ratio of calculated to experimental values) were rarely close to 1. If attention is concentrated to the pressurizer and the iodine results are neglected, the best agreement was obtained with RETAIN-2c, RAFT, and the Italian calculations with TRAP-MELT. For RETAIN-2c it was, however, observed that the large particle sizes which lead to the good agreement were due to a coding

error in the program. The relatively good agreement with TRAP-MELT (Italy) was caused by a division of the pressurizer into three parts separated by horizontal planes. Each section had the sedimentation area of the bottom plane. This method can at least be said to be controversial. The good agreement with RAFT was due to the large particle diameters obtained in the nucleation calculation. The wall deposition was underestimated by all codes.

In the piping the sedimentation was overestimated while the vertical surface deposition was usually underestimated. The discrepancies could be due to errors in the calculation of turbulent deposition or to the effect of relocation/reentrainment. A part of the material which originally had deposited in the pipes might have been moved to other zones, by gravitation or resuspension.

The underestimation of deposition on the vertical piping might be due to neglecting wall roughness, besides relocation. This is one area where more development work should be done.

Test 2a was performed 1983-05-03. The performance of the test fell short in several respects. Problems were experienced with the plasma arc heaters, with steam flow, with feeding of tellurium and with sampler filter performance. The results could therefore hardly be used for code validation.

Test 2b was performed 1984-01-17 and was a more successful repetition of test 2a. The calculational results diverged greatly between the different codes and also between the different TRAP-MELT calculations. The C/E values were rarely close to 1. If attention is concentrated to the pressurizer the best agreement was obtained with RAFT, PATRAP, and RETAIN-2c. For RETAIN-2c it must again be observed that the large particle sizes which lead to the good agreement were due to a coding error in the program. The relatively good agreement with PATRAP is probably due to the very special method for calculating the particle sizes. The good agreement with RAFT was again due to the large particle diameters (MMD= 4 μm) obtained in the nucleation calculation. The measured AMMD was about 11 μm, that is MMD=about 5 μm. The TRAP-MELT results at BCL were AMMD=4.4 μm.

For the partition of deposition between floor and wall the PATRAP code again underestimated the wall deposition while the French results were the opposite. Williams and Butland (WIL84) proposed an investigation of the reason why most codes underestimate the wall deposition. According to a private communication, they now believe that they have an explanation.

Recently, Parozzi et al. (PAR88) have improved TRAP-MELT as regards the wall deposition effect. By considering centrifugal forces from gas circulation the ratio of wall-to-total deposition increased from 0.81% to 10.6%. The experimental value was 12.5% .

In the piping the sedimentation was overestimated while the vertical surface deposition was usually underestimated. The phenomenon seems to be the same as in test 1.

Test 7 was performed 1984-11-07 and was the last fissium test. The complete facility, including the reactor vessel, was now used.

In the calculations presented at meetings, difficulties were encountered due to the high temperature in the reactor vessel. No fissium aerosol could be formed in the lower part but some codes accounted for chemisorption. The discrepancies between the measured and calculated values were generally very large and they seem to be due to the difficulty of predicting the partition between vapour and aerosol and calculating the interaction between vapour and surfaces. The percentage of vapour will of course also have an effect on the particle size distribution.

Downstream of the reactor vessel Williams (WIL86) had assumed a fissium source rate as determined from the experimental mass balance. His results for these volumes with an initial AMMD of 8 microns lead to better agreement between experiments and calculations than that obtained in the reactor vessel, at least for the pressurizer and the horizontal pipes. The differences between the different TRAP-MELT results could be partly dependent on different input data.

The largest discrepancies downstream of the vessel were encountered in the pipes. This has sometimes been thought to be due to problems with modelling deposition in bends.

Test 4 was performed 1985-02-27 and was the only test with corium. The complete facility was used. The test was reasonably successful in achieving the planned goals.

In the calculations presented at meetings, difficulties were encountered due to the high temperature in the reactor vessel. No fissium aerosol could be formed in the lower part but some codes accounted for chemisorption. Williams (WIL86) has studied the influence of variation of the ratio between the vapour and the particle parts of the fissium source and of the particle shape factors. The initial particle diameter was an independent variable. The deposition was calculated as a function of initial particle size.

The deposition in the reactor vessel was for CsOH mainly due to chemisorption. Models for chemisorption occurred only in TRAP-MELT and RAFT. Agreement in vessel deposition between experiment and calculation was obtained only for RAFT. Moreover this agreement was only fortuitous, because it did not hold for the different parts of the vessel. It is also somewhat perplexing that the deposition of cesium and silver had about the same rates although cesium cannot condense in the lower part of the vessel.

Downstream of the reactor vessel the discrepancies were generally large, but the most consistent results were again obtained with RAFT. The differences between the different TRAP-MELT results could be partly dependent on different input data.

Fig. 4.3.1 illustrates the very different results obtained with the codes used in 1986.

The most recent improvements in calculational methods were reported by Williams at a meeting in Montreux (WIL88). He has developed a model for "intervolume gravitational coagulation" in the VICTORIA code and with this model he obtained good agreement between experiments and calculations for all volumes, including the pipes.

To summarize, the severe test conditions in Marviken-ATT limited the experimental methods and also the accuracies, but important conclusions can still be drawn, for example:

- The deposition in pipe bends is important and the theoretical models for these mechanisms should be improved.
- In 1986 the prediction of deposition in horizontal pipes was in error by at least a factor of 2-3. The deposition on vertical surfaces was underestimated. In order to solve these problems surface roughness should be considered.
- The large particle sizes can probably be calculated if nucleation is considered, but it is also possible to use by the intervolume settling model implemented in VICTORIA-UK.
- Models for chemisorption of tellurium on stainless steel have to be improved.
- Models for chemisorption of cesium hydroxide on stainless steel have been improved at AEEW.
- In connection with the development of models for chemisorption, small scale experiments could and should be done.

TABLE 4.3.1

CHARACTERISTIC FEATURES OF THE AEROSOL CODES

B = bin model for aerosol size distribution
L = log-normal aerosol size distribution
P = Pruppacher-Klett formula for collision efficiency
F = Fuchs formula for collision efficiency
BD = Calculation of deposition in bends
CE = Calculation of chemical equilibrium
CHS = Calculation of chemisorption
N = Calculation of nucleation
VCE = Consideration of vapour condensation and evaporation

Code Code	Size dist.	Coll. eff.	BD	CE	CHS	N	VCE
AEROSOLS/B1	B	P	yes	no	no	no	no
MAAP-3	-	F	yes	no	no	no	yes
PATRAP-3	B	P	no	yes	no	no	no
RAFT	B	P	no	yes	yes	yes	yes
RETAIN-2c	L	P	no	no	no	no	yes
RETAIN-S	L	P	yes	no	no	no	yes
TRAP-MELT-2	B	P	yes	no	yes	no	yes
TRAP-MELT2-UK	B	P	no	no	yes	no	yes
TRAP-MELT2/ENEL	B	F	no	no	yes	no	yes

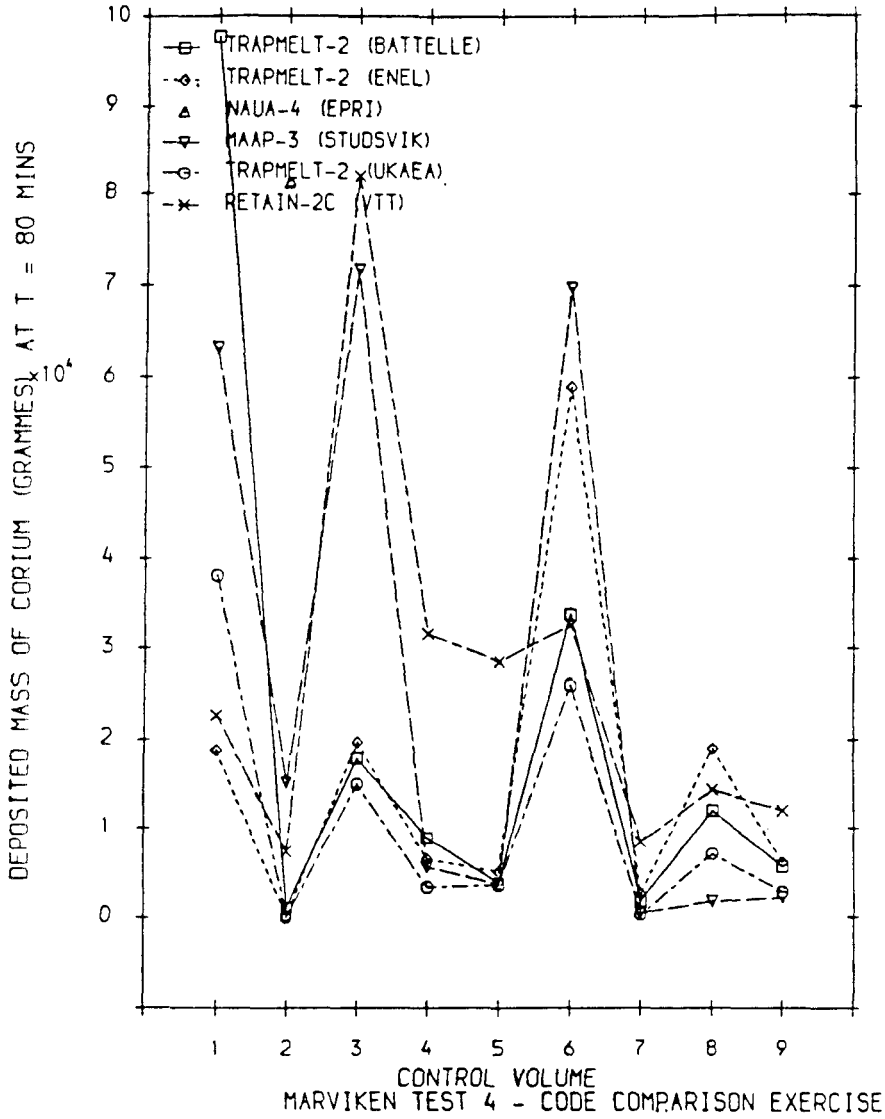

Fig. 4.3.1 Results obtained for the Marviken Test 4 in code comparison calculations reported 1986.

- The effect of hygroscopicity of cesium hydroxide has been considered at Harwell (Knights). This can be a source of relocalization of deposited material.

- The different aerosols seem to have been transported and deposited separated from each other only in the vessel. The question of whether such a separation is necessary in the codes should be discussed.

- Until 1987 the codes used for test predictions were not able to give an agreement, even within a factor of two, for the deposition in any compartment if all tests were considered, even if the experimental uncertainties were considered. In particular, the phenomena in the reactor vessel where the temperature was about 1000 °C could not be modelled by the codes. The importance of modelling the fission product transport in the reactor vessel should perhaps be made the subject of a more serious discussion.

4.3.2. DEMONA Benchmark Exercise

The DEMONA experiment B3 was selected for a benchmark exercise with participation from the EEC countries, Sweden, and the United States. From the Nordic countries Studsvik Energiteknik AB and Risoe National Laboratory took part. For a detailed account, see SCH88.

The data supplied were all experimental - geometric data, thermal-hydraulic data, aerosol source rate etc. The only extra help was provided by the steam condensation rates as calculated by the KWU code COCMEL.

Results

In the exercise 6 different codes were used, one of these (NAUA) in 6 different versions. In general the results agree closely with each other, especially on the three important parameters airborne, leaked and settled masses. Comparison shows that the leaked mass results of the single-volume, discrete particle size distribution codes agree within a factor of 2 (Figs 4.3.3).

Generally, comparison with experiment is also satisfactory (Figs 4.3.2 and 4.3.3). Most of the results are moderately conservative throughout, but two codes underestimate the peak value of the mass concentration by perhaps 25%, probably owing to an overprediction of sedimentation. For the MAAP results, see paragraph 5.1. There is also a considerable scatter at long times of the non-NAUA calculations, reflecting the different bases of the codes.

It was also found that the results were sensitive to the thermal-hydraulic input, especially to the quantities that influence the condensation. From this it may be concluded that improvements are needed more in the thermal-hydraulic codes and in the coupling to the aerosol codes than in the aerosol codes themselves.

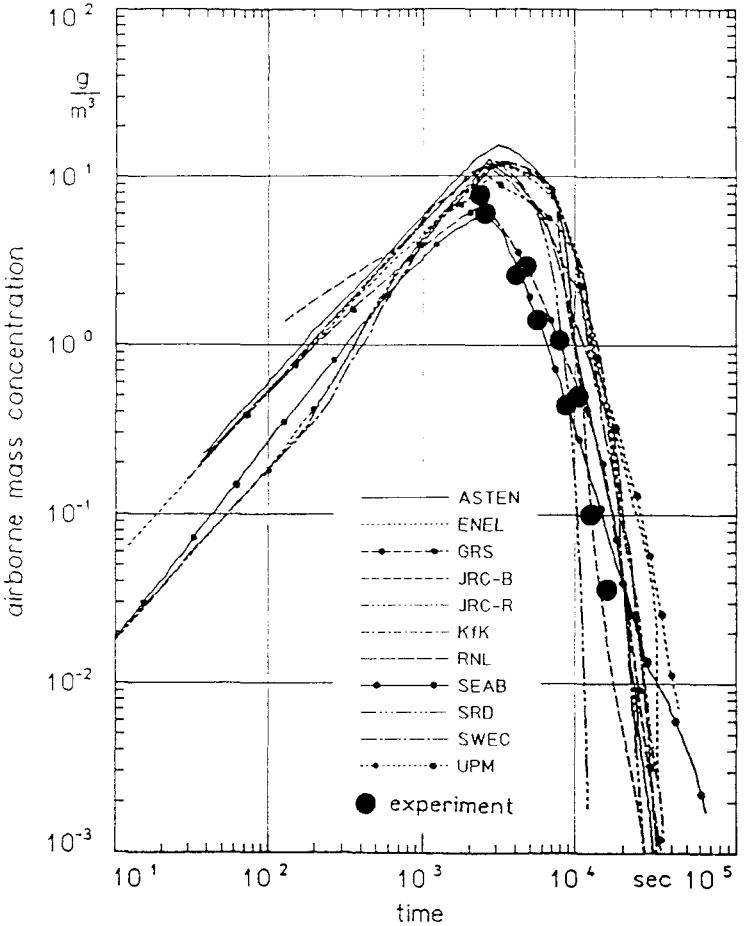

Fig. 4.3.2 DEMONA experiment B3. Calculated airborne mass concentrations compared to experimental results. (SCH87).

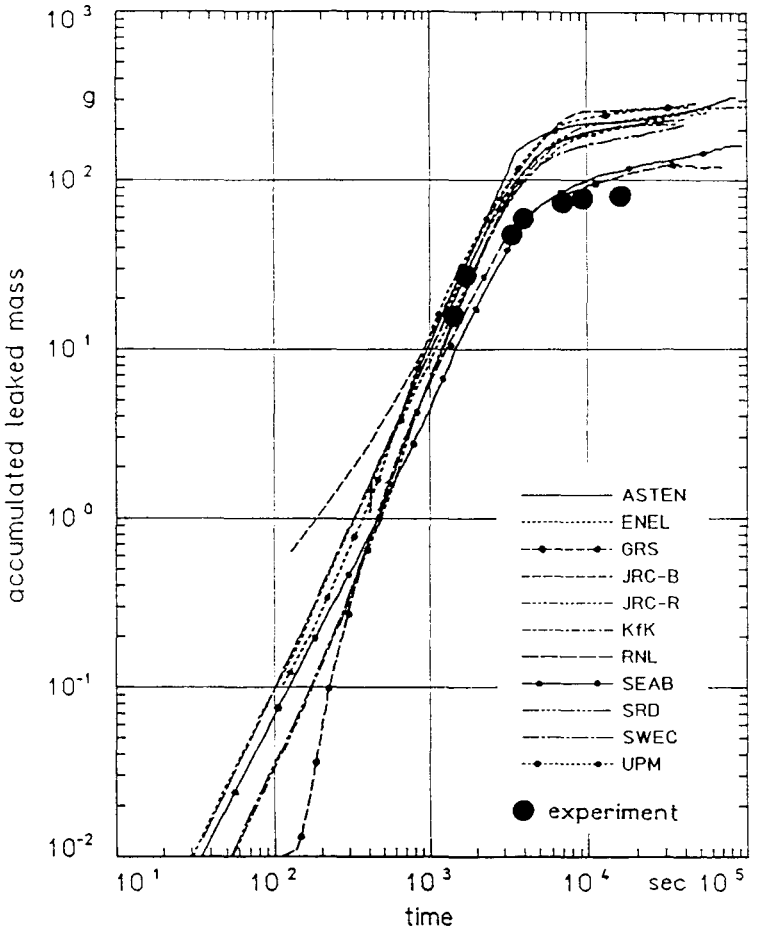

Fig. 4.3.3 DEMONA experiment B3. Calculated leaked masses compared to experimental results. (SCH87).

4.3.3 LACE

For most tests pre- and posttest code comparisons were carried out. For pipe tests LA1, LA3A, LA3B and LA3C the most important aerosol removal processes were turbulent deposition and deposition in pipe bends. In Table 4.3.2 (Ref WRI88) there is a description of the deposition models used in codes participating in the LA3 code comparison exercise.

In the the blind posttest analyses the last version of TRAP-MELT had the best capability to simulate particle deposition in the LA3 test. Later a similar model for turbulent and bend deposition was included in the RAFT code. It is thought that in the LA3C test the discrepancies between TRAP-MELT and test results are caused by translocation of deposited wet slurry ($CsOH+H_2O$(liquid) -- MnO(solid)). None of the codes had a model for this translocation process.

In the containment tests codes having a model for the behaviour of hygroscopic particles (NAUA-HYGROS, VTT/EPRI and NAUA, Stone & Webster) had the best ability to simulate aerosol behaviour. In the LA4 test different behaviour of CsOH and MnO was observed, because of overlapping injection periods. This phenomenon could best be predicted by aerosol models with hygroscopic multicomponent models (NAUA-HYGROS, EPRI and NAUA, St&W).

There was in general good agreement between the codes modelling dry aerosols. Fig. 4.3.4 shows results from pretest calculations on LA4 with different codes. There was unfortunately a coding error in the MAAP-3 module used. Results with a corrected version are given in section 5.3.

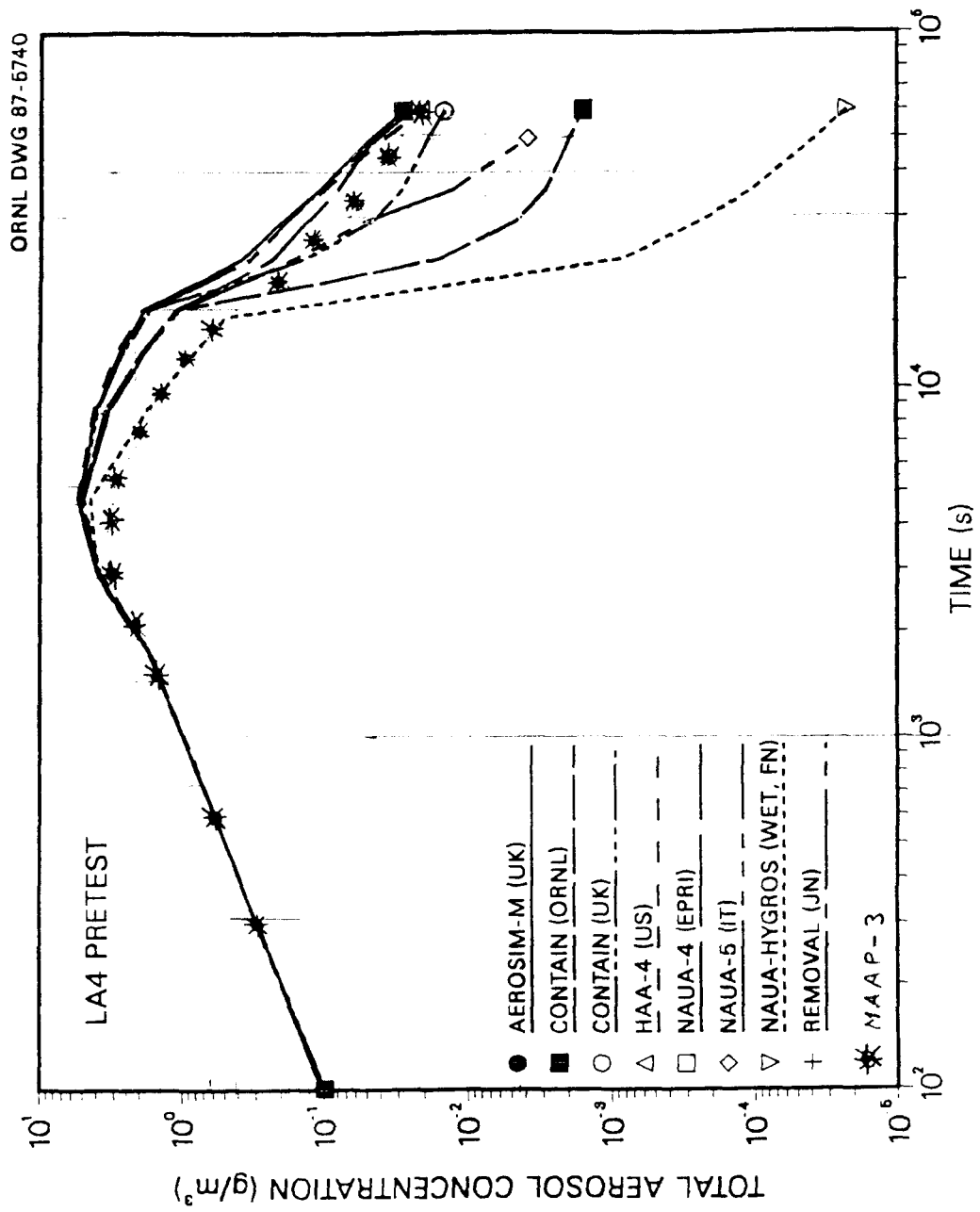

Fig. 4.3.4 Results from pretest calculations on LACE - LA4 with different codes.

4.4 Conclusions from two international symposia

In two international symposia the recent state of the art has been summed up. The first of these was the "WORKSHOP ON WATER-COOLED REACTOR AEROSOL CODE EVALUATION AND UNCERTAINTY ASSESSMENT" held in Brussels in September 1987 and organized by the OECD. As examples of areas where significant progress was reported the following can be mentioned:

* systematics in physical models
* independent transport of different species
* deposition in pipe bends
* effects of turbulence
* effects of hygroscopicity
* nucleation

An important part of the meeting was devoted to the analyses of the LACE and DEMONA experiments. From the Summary conclusions the following is cited about these experiments:

"They have given direct evidence of the importance of coupling of aerosol behaviour to thermal-hydraulics, and of chemical effects such as hygroscopicity.------
The basic theories required to handle the key phenomena are now well developed, and the remaining requirements centre on improved input data , and some outstanding aspects on the implementation of the theoretical models in computer codes."

Other conclusions made were:

* "The well-mixed assumption on which many codes are based may be inapplicable to large containment structures."

* "Certain of the large-scale experiments provide evidence of inhomogeneities, and suggest that natural convection effects can be important."

* "On -- aerosol shape factors and leak path retention, little or no progress was apparent --."

* "The incorporation of mechanistic models to handle the diverse aspects of thermal-hydraulic and chemical interactions is leading to aerosol codes of increasing complexity ---."

* "Uncertainty assessment --- is clearly a topic meriting considerably more attention."

The second meeting was the "MARVIKEN V/DEMONA/LACE WORKSHOP" held on June 29 to July 1, 1988, in Montreux. Some more progress was reported during the preceding year, nota bene concerning the following phenomena:

* the effect of gas circulation

* inter-volume coagulation
  This is a model developed for VICTORIA by Williams (WIL88) which divides a volume horizontally into a number of nodes with different particle size distributions. Gravitational coagulation is calculated separately for each node and the vertical flow and particle transport between nodes are considered.

* chemisorption

The uncertainties were concluded to be smaller in containment calculations than for the primary circuit, much depending on the different chemical conditions. The need for small scale experiments was recognized. Swedish representatives declared that protection and safety measures must be such that the present uncertainties can be tolerated.

5. RESULTS OF WORK IN THE NORDIC AEROSOL TRANSPORT PROJECT

5.1. Code Comparison

In the Nordic countries three types of containment aerosol codes have been used: Moment-generating codes (RETAIN-S and HAARM-S, whose aerosol parts are almost identical), a "bin" code (NAUA), and a "scaling" code (MAAP). The differences between the three types are closely connected to their descriptions of the aerosol size distribution.

In NAUA the size distribution is described by a number of size classes, the number of aerosol particles in each class being the variables (typically 20-50 variables). The aerosol processes are readily formulated and the code is flexible and gives detailed information (BUN83).

Several versions exist; the newer ones are usually based on NAUA/5 (BUN87). The main difference between NAUA/5 and NAUA/4 is that NAUA/5 can treat diffusiophoretic deposition. In the comparisons with other codes NAUA was the reference code.

In moment-generating codes the size distribution is required to have a predescribed mathematical form throughout the calculations, usually the log-normal distribution. The main variables are the parameters of the distribution: position, width, and area under the distribution graph, or equivalently the zeroth, first and second moments of the distribution. The aerosol processes require a certain amount of analytical work before implementation. Distributions which deviate much from the log-normal are not well described by codes of the HAARM family. Examples include the two-hump distribution resulting from steam condensation on particles, and distributions of particles that initially have different densities. The advantage of these codes is that they are very fast because very few differential equations have to be solved. The results are not precise, but they can be used in combination with codes like NAUA for parameter studies.

The scaling code MAAP does not consider the size distribution explicitly. Instead it applies empirical correlations for quantities such as suspended mass concentration, particle size, and time, which are nondimensionalised by scaling. The correlations are derived from experiments and from model calculations with more detailed codes. The basis is the assumption that for a dense aerosol agglomeration will soon transform the initial size distribution into a size distribution characteristic of the suspended mass concentration. Different sets of correlations for the various mechanisms apply for the two cases, continuous source and aging aerosol. MAAP allows fast computations, but if two sources have different

material densities or if steam condenses on the particles the correlations run into trouble (FYN88, EPS88). A detail which probably has some importance for the MAAP calculations made is that the shape factors are fitted to LMFBRs instead of LWRs. They should be changed to 1 in the parameter file.

Calculations show the following differences between NAUA and HAARM (or RETAIN) (FYN85, HÄG85):

- When the source is "on", HAARM overestimates the suspended mass.

- Some time after the source has ceased, HAARM overestimates the decay rate of the aerosol. This may easily result in a whole order of magnitude underprediction of the suspended mass. Only part of this discrepancy results from the incorrect use in HAARM of the Fuchs collision efficiency for gravitational agglomeration (NAUA applies that of Pruppacher and Klett) (LAU85b).

Comparison of MAAP and NAUA suggested (HÄG86, FYN88, SCH88) that:

- MAAP overestimates deposition when the source is "on".

- MAAP has difficulty in calculating "wet" aerosols.

However, it was later recognized that some of these errors were caused by using the module for the auxiliary building, AUX, instead of the subroutine for the primary circuit and containment. The AUX module only considers the model for aging aerosols. It has been changed at VTT to include the model for continuous source and new calculations have been made with this version (HÄG89). Some discrepancies still remained between MAAP-AUX and NAUA, particularly in a two-compartment case simulating a loss of power accident sequence for the Oskarshamn 2 reactor. The compartments represented the reactor vessel and the containment. In NAUA calculations a source from core-concrete interaction was assumed in the containment, in addition to the source in the vessel. MAAP-AUX could only consider the source in the vessel. The results are given in figs 5.1.1, 5.1.2 and 5.1.3 for aerosol concentration, deposition and leakage, respectively, of the inert material in the containment. The core-concrete interaction started at 10.800 s and comparison of the results after this time is therefore difficult. It can, however, be concluded that MAAP-AUX overestimates the deposition even at earlier times, possibly depending on the strongly varying source in the vessel. The effect is probably cumulative when the number of compartments increases. It should be pointed out that the hygroscopicity effect is not considered in this case.

This effect is, however, considered in the archived MAAP version and can cause still higher deposition. Comparisons between NAUA and HAARM are shown in figs 5.1.4 to 5.1.6.

OSKARSHAMN 2, sequence AB
Inert aerosol concentration in containment

Full line: MAAP-AUX
Slashed line: NAUA

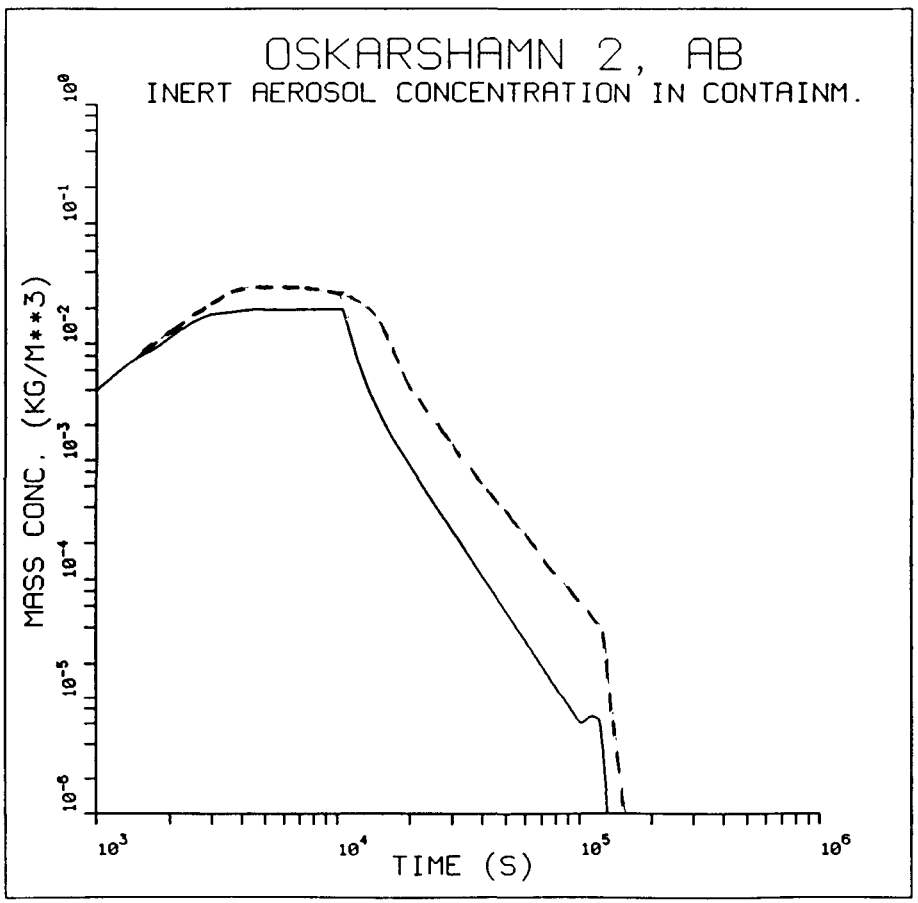

Fig. 5.1.1

OSKARSHAMN 2 , sequence AB
Deposited inert mass in containment

Full line: MAAP-AUX
Slashed line: NAUA

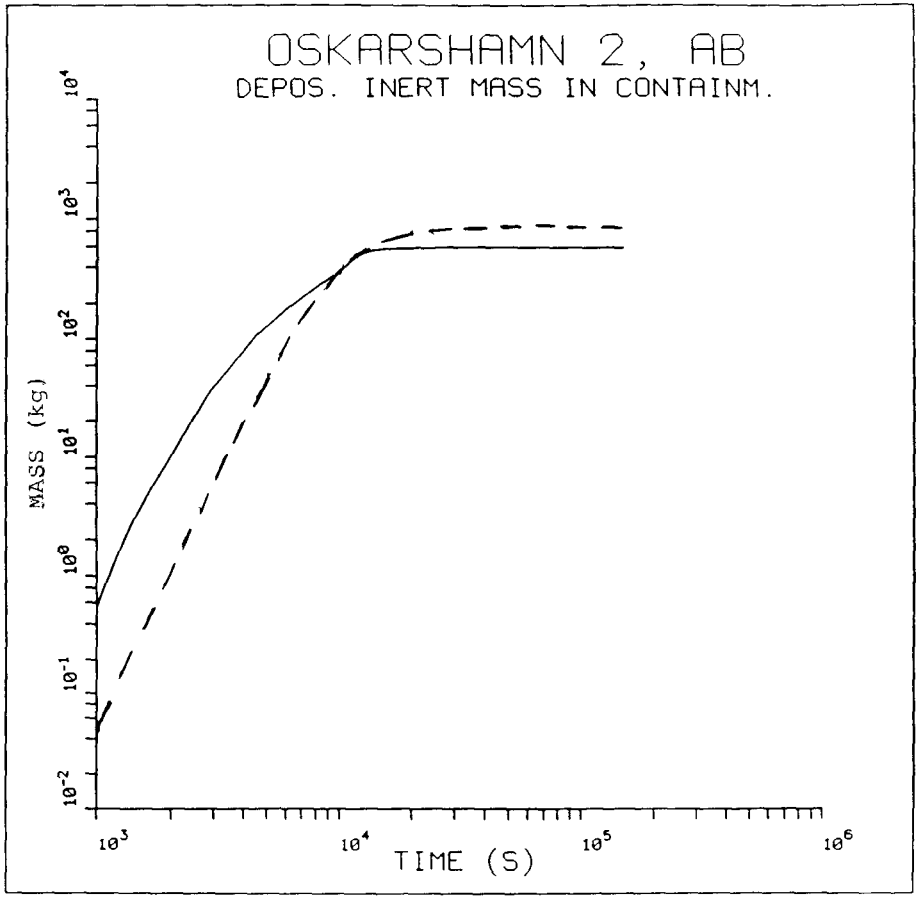

Fig. 5.1.2

OSKARSHAMN 2 , sequence AB
Leaked inert mass from containment

Full line: MAAP-AUX
Slashed line: NAUA

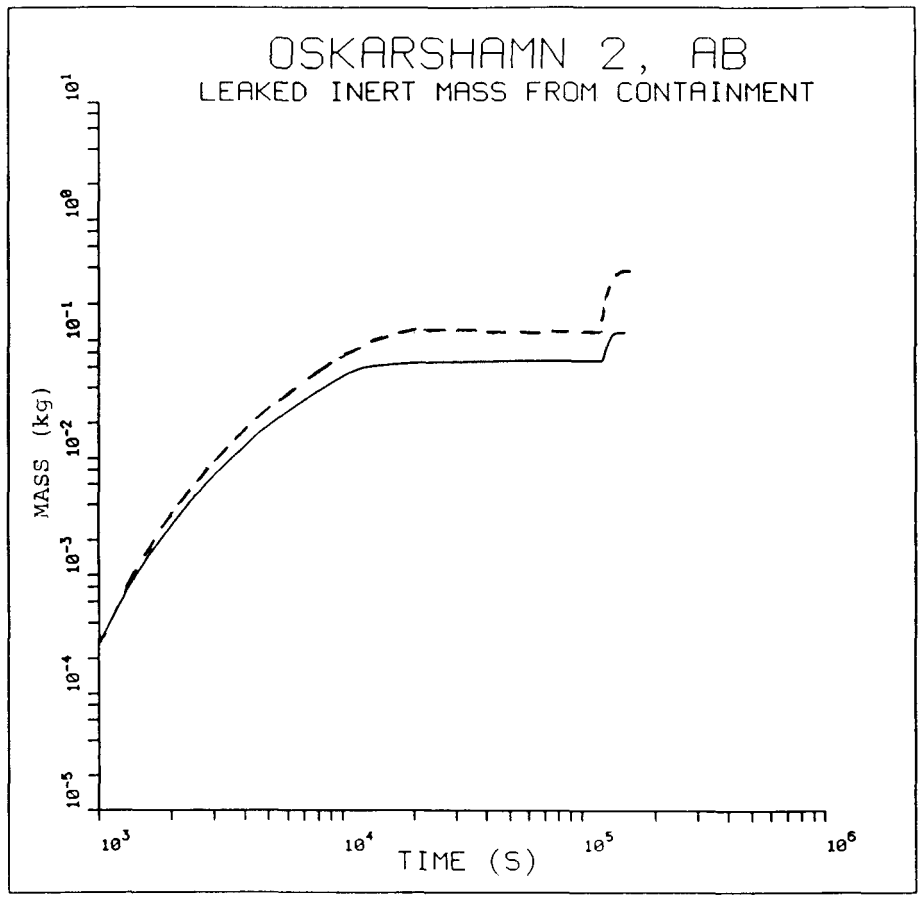

Fig. 5.1.3

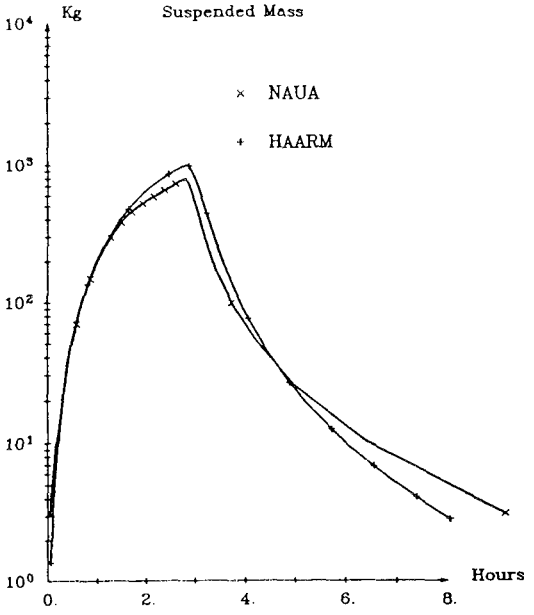

Fig. 5.1.4

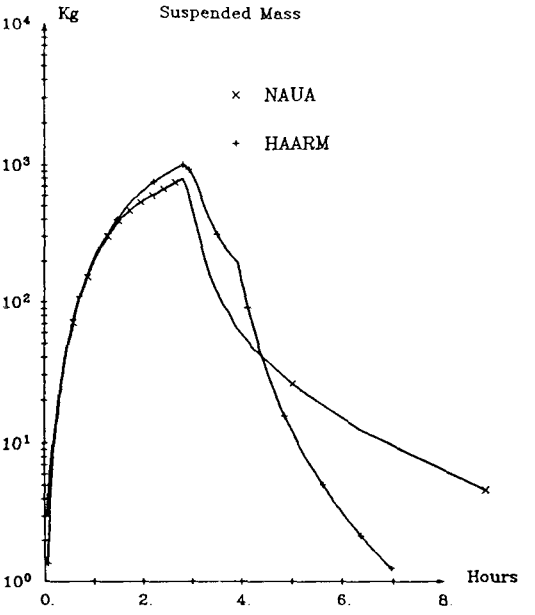

Fig. 5.1.5

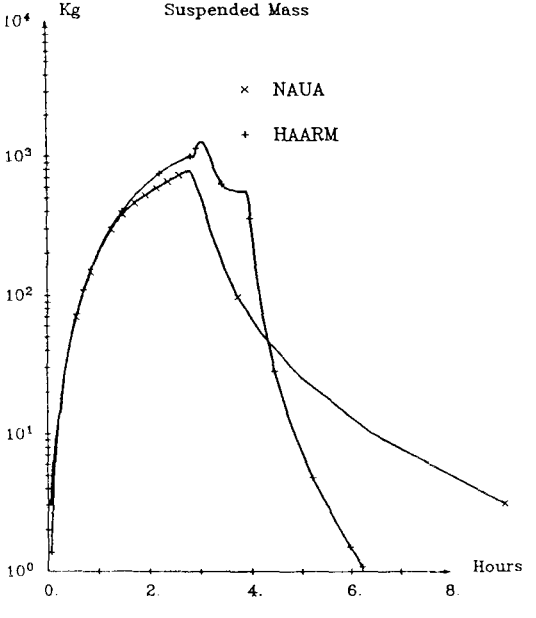

Fig. 5.1.6

5.2. Aerosol Nucleation and Condensation

5.2.1. Phenomenology

A supersaturated vapour may undergo a phase change to form an aerosol in two ways: by homogeneous nucleation and by heterogeneous condensation. Homogeneous nucleation means that nuclei are generated in a homogeneous phase. This excludes ions, dust, and wall surfaces. Heterogeneous condensation takes place on existing particles without the formation of new nuclei.

In this section the classical theory of homogeneous nucleation will be considered from the points of view of thermodynamics and kinetic gas theory. Only a qualitative account of the classical theory will be given, as this gives the essential physics and suffices for an understanding of the main results. After that, heterogeneous condensation will be considered briefly.

5.2.1.1. Homogeneous Nucleation

Homogeneous nucleation results from density fluctuations of the vapour, which produce microscopic clusters consisting of relatively few molecules, of the order of 100 or less. Clusters below a critical size are unstable; above that size they become stable.

Consider the formation of one cluster with radius r in a supersaturated vapour. The saturation ratio S is the ratio between the actual vapour pressure and the equilibrium vapour pressure at the temperature T.

The change of the Gibbs free energy, G, consists of two terms. One is proportional to volume and negative because the bulk chemical potential of the liquid by definition is lower than that of the supersaturated vapour. The other term is positive. It is the product of surface area and surface tension. For small r, G increases because the surface term is proportional to r^2 whereas the first term is proportional to r^3. For large r the change in G is negative.

The maximum increase, G_c, in G is obtained at the critical radius r_c, which marks the border between unstable clusters ($r<r_c$) and stable droplets ($r>r_c$).

G_c can be regarded as the height of a barrier that the system as a whole must surmount to create one droplet.

An analysis shows the expected result that both r_c and G_c decrease with increasing S. For instance, G_c is inversely proportional to $(\ln S)^2$.

If the nucleation rate is required, then thermodynamics must be supplemented with kinetic theory.

In the kinetic theory condensation and evaporation are treated as a series of processes of the type

$$A_g + A = A_{g+1} \qquad \text{(or the reverse)}$$

in a chemical notation where g is the number of molecules in the cluster A_g. The impingement rate of the "A" molecules upon A_g is calculated from kinetic gas theory. The calculations are lengthy but the result is relatively simple: The nucleation rate density is obtained in the form

$$J = f.n^2.\exp(-G_c/kT).$$

Here, the factor f is almost constant and n is the concentration of vapour molecules. The exponential varies strongly due to the dependence of G_c on S. An increase by 10 orders of magnitude or more when S increases from 2 to 3 (UO_2, KEN77) or from 3 to 4 (Water, FRI77) is not uncommon.

Mention is made at this point of heterogeneous nucleation, in which a solid substrate takes part. The treatment follows that of homogeneous nucleation in many respects and the results are qualitatively similar (existence of a critical size etc.). However, the different natures of the systems must be kept in mind. One important complication is that the nucleating phase in many cases is the adsorbed layer of vapour molecules on the substrate. The barrier G_c and the nucleation rate depend on the shape of the cluster and the contact angle and a line tension may be important in addition to the surface tension. For LWR safety heterogeneous nucleation is important because of steam condensation on aerosol particles in the containment. Fortunately it is most naturally treated in the codes as heterogeneous condensation.

5.2.1.2. Heterogeneous Condensation

When supersaturation is low and a large concentration of particles is already present, condensation takes place on the existing particles and no new nuclei are formed.

For particles much smaller than the mean free path, l, of the vapour molecules, kinetic theory must be used. The condensation rate is then found to be proportional to r^2 and to (p-p(r)), where r is the particle radius, p is the

vapour pressure and p(r) is the pressure in equilibrium with a droplet of radius r. In the continuum range diffusion theory gives condensation rates proportional to r and to (p-p(r)). An approximate interpolation formula between the two extremes is given by Fuchs and Sutugin (see FRI77):

$$F = 4\pi Dr(p-p(r))(1+Kn)/(kT(1+1.71Kn+1.333Kn^2))$$

F is the condensation rate, D is the vapour diffusion coefficient, and Kn is the Knudsen number, Kn = l/r.

5.2.1.3. Discussion

The model given above of homogeneous nucleation is qualitatively correct. It has been criticised for a number of reasons, however. Two of these are latent heat, which is not included in the analysis, and the impossibility of extrapolating macroscopic values of specific volume and surface tension to clusters consisting of 20 - 100 molecules.

Nevertheless, the predictions of critical saturation ratios (one nucleus formed per cub.cm per sec.) are remarkably good (GRE64), especially when latent heat is included (STR86), reflecting the fact that the physics is essentially correct.

For condensation, more advanced models than that of section 5.2.1.2 have been developed and implemented in codes. In the NAUA code Mason's equation is applied to the condensation of steam. The RAFT code applies an approximation, in which the gas surrounding the particle is divided into two layers, where the inner layer of one mean free path thickness is treated in some detail. Both of these codes include the effect of latent heat. The code TRAP-MELT3 will be considered below.

5.2.2. Aerosol Nucleation in TRAP-MELT3

5.2.2.1. Heterogeneous condensation

The model for condensation and evaporation of fission products in TRAP-MELT3 is simple. The transfer rate between vapour and condensate on walls or particles is proportional to the area available and to the difference between the vapour concentration in the gas and the vapour concentration in equilibrium at the temperature given (JOR85). Aerosol particles are assumed to have the temperature of the gas, whereas the walls may have individual temperatures. The mass transfer coefficients (the proportionality constants) are given by elementary correlations in view of the data available and of the accuracy of the code in general. For the wall mass transfer turbulent pipe flow is assumed and for the particle mass transfer simple diffusion is assumed.

The equations of the model are treated separately from the equations describing the transport, removal and agglomeration. This is done partly because the processes of condensation and evaporation are so rapid that simultaneous solution would introduce stiffness into the equations and partly because of the difficulties that arise when the aerosol mass or the condensed mass on the walls vanishes because of evaporation.

Considering the other approximations the whole procedure is reasonable. It does have some weaknesses, however. First, simple diffusion will overestimate the mass transfer for small particles. Second, the particle surface area available for condensation or evaporation is kept fixed during a time step, although it varies - in certain cases strongly - during the time step owing to the mass transfer. And finally, the amount of vapour added or removed by gas flow is introduced as a "puff" at the beginning of each time step, not continuously.

5.2.2.2. Homogeneous nucleation

If the fission product vapour is supersaturated at the end of a time step, the surplus vapour is assumed to condense with equal masses into the three lowest size classes. This "homogeneous nucleation" is of course arbitrary with respect to the condensed mass, to the number of size classes involved, and to the distribution amongst these classes. The details can be discussed but in view of the available data and the fact that the small particles will soon take part in agglomeration, thereby wiping out the details of their initial distribution, the procedure seems reasonable.

5.2.3. Aerosol Nucleation in RAFT

The temporal and spatial change in particle size spectrum as a result of homegeneous nucleation, heterogeneous nucleation, agglomeration, and deposition is described in the RAFT code by the population balance equation (IM85):

$$\frac{\partial n}{\partial t} + \frac{1}{A}\frac{\partial}{\partial x}[Aun] + \frac{\partial}{\partial r}[\dot{r}n] = \sum_{j=1}^{m} J_j \delta(r-r_j^*) + \left[\frac{\partial n}{\partial t}\right]_{coll} - v_d An$$

(a) (b) (c) (d) (e)

where n(r,x,t) is the particle size (r) distribution function at position x and time t. The physical interaction terms correspond to: (a) convection of particles (particle-bulk); (b) particle growth rate due to heterogeneous nucleation (particle-vapour); (c) rate of particle formation of critical size r* due to homogeneous nucleation (vapour-vapour); (d) particle collision and attendant agglomeration (particle-particle); and (e) rate of particle removal due to deposition on boundary surfaces (particle-surface).

Homogeneous Nucleation

In a rapid expansion or cooling process, the chemical potential of a vapour can become higher than its equilibrium value. This unstable situation manifests itself by the partial pressure of a constituent exceeding its own saturation pressure. The saturation ratio is:

$$S = \frac{p_v(T)}{p_s(T)} \quad , \qquad \begin{cases} > 1 & \text{supersaturation} \\ = 1 & \text{saturation} \\ < 1 & \text{subsaturation} \end{cases}$$

A decrease in supersaturation towards unity reflects the approach of a system to a thermodynamically favoured state. This is accomplished through a phase change, that is, the vapour molecules nucleate into aerosol particles.

In classical nucleation theory, the rate expression is derived by considering a series of reactions between the monomer (vapour molecule) and clusters (collection of n molecules also called n-mer). The reactions involve addition of a monomer to the cluster (condensation) or removal of a monomer from a cluster (evaporation). Reactions between the clusters are neglected since their concentration is much smaller than that of the monomers.

The nucleation kinetics are described in terms of the equilibrium distribution of the clusters. The Gibbs free energy of formation of a cluster from the vapour state is determined by the balance between the surface tension energy of the droplet and the free energy change in the condensation process. The Thomson-Helmholtz equation provides a thermodynamic relation for critical cluster size which maximizes the change in Gibbs free energy (IM85):

$$r^* = \frac{2\sigma v_m}{kT \ln S} \quad ,$$

where σ is the surface tension and v_m the molecular volume in the condensed state. Droplets smaller than this size are not stable but droplets larger than r* will grow by condensation and agglomeration.

The following expression for homogeneous nucleation rate is used in RAFT:

$$J = \beta(4\pi r^{*2}) Z N \exp\left(-\frac{\Delta G}{kT}\right) \quad ,$$

where β is the specific impingement rate of monomers with a flat surface,

$$\beta = \frac{p(T)}{(2\pi m kT)^{1/2}} \quad .$$

Z is the Zeldovich factor,

$$Z = \frac{V_m}{2\pi r^{*2}} \left[\frac{\sigma}{kT}\right]^{1/2} \quad .$$

N is the concentration of monomers,

$$N = \frac{P}{kT}$$

ΔG is the change in Gibbs free energy.

$$\Delta G = (4\pi/3) r^{*2} \sigma .$$

In RAFT there is also a possibility to take into account the surface tension dependence on particle radius and a correction factor caused by particle vibration. However, the experimental quantification of these factors is still uncertain.

It should be noted that nucleation on ions produced by radiation can also be considered in the RAFT code.

There are several uncertainties associated with the rate of particle formation by homogeneous nucleation. However, these uncertainties have only a secondary importance as compared to particle growth by heterogeneous nucleation and the input thermodynamic parameters, when aerosol transport in the primary system is evaluated.

Heterogeneous Nucleation

In the RAFT code the phenomenology used to describe heterogeneous nucleation is very similar to that of steam condensation on particles (detailed description in section 5.3.1). The final equation for the mass flux towards a particle differs from that derived in section 5.3.1 (Mason eq.) only as a result of using slightly different approximation methods. In the RAFT code the following set of equations is used to calculate the mass flux to the particle (for explanation of symbols see nomenclature) (IM85):

$$\dot{m}_j = (4\pi r D_j) f_j \left(\frac{p_{sj}}{R_j T_g}\right) \left\{ S_j - \left[1 + \left[\frac{L_j}{R_j T_g} - \frac{g}{2(Kn_g+1)} - \frac{\Phi_j}{kT_g} \right] \frac{\Delta T}{T_g} \right] \exp\left[\frac{\Phi_j}{kT_g} \right] \right\}$$

$$\frac{\Delta T}{T_g} = \frac{\Sigma\, a_j \left[S_j - \exp\left(\frac{\Phi_j}{kT_g}\right) \right]}{1 + \Sigma\, a \left[\frac{L_j}{R_j T_g} - \frac{g}{2(Kn_g+1)} - \frac{\Phi_j}{kT_g} \right] \exp\left(\frac{\Phi_j}{kT_g}\right)}$$

where

$$a_j = \frac{L_j f_j D_j p_{sj}}{g k_g R_j}$$

Φ is the energy function defined as the work required to construct a spherical droplet from the volume of the bulk liquid.

$$\Phi_j = \frac{2\sigma_j m_j}{\rho l j^r}$$

Kn_j is the Knudsen number for the condensing vapour.

$$Kn_j = \lambda_j / r$$

$$R_j = k/m_j$$

and

$$\frac{1}{f_j} = \left[\frac{1}{(Kn_j+1)} + \left[\frac{\alpha_j D_j}{r} \right] \left[\frac{2\pi}{R_j T_b} \right]^{1/2} \right]$$

$$\frac{T_b - T_g}{T_s - T_g} = \frac{g}{Kn_g + 1}$$

where

$$\frac{1}{g} = \left[\frac{1}{(Kn_g+1)} + \left[\frac{D}{r} \right] \left[\frac{2\gamma}{\gamma+1} \right] \left[\frac{2\pi}{R_g T_g} \right]^{1/2} \right]$$

and $\Delta T = |T_s - T_g|$.

In Table 5.2.1 there are results of particle size and number concentration after nucleation for some accident sequences and experiments simulated with the RAFT code (IM87).

Table 5.2.1

SUMMARY OF SOME RAFT SIMULATIONS

Transient	Nucleating Species	P (atm)	u (m/s)	x_j (ppm)[a]	$u \frac{dT}{dx}$ (K/s)	Approx. N (#/cm^3)	Approx. $\langle r \rangle$ (μm)	Application
AD	CsI	3.7	0.55	443	1618	10^7	0.5	STEP[b]
TQUW	CsI	1.4	1.26	209	3408	10^8	0.1	STEP
TMLB'	CsI	81.6	0.02	207	44	10^5	5	STEP
TMLB'+C	Ag	81.6	0.03	301	97	10^6	5	STEP
S_2D	CsI	100.	0.15	840	1.42	10^2	30	PWR
MARV 1[c]	CsOH	1.08	0.98	65,220	1347	10^5	4	MARVIKEN[d]
MARV 2A[c]	CsI	1.08	1.25	8,802	1136	10^5	3	MARVIKEN
MARV 2B[c]	CsI	1.08	0.94	8,590	803	10^5	4	MARVIKEN
MARV 4	Ag	1.08	4.30	81,590	5303	10^7	6	MARVIKEN
MARV 7	CsI	1.08	0.46	2,689	84	10^4	5	MARVIKEN
BI	CsI	1.0	2.15	277	342	10^7	0.1	HOT TUBE[e]
COH	CsOH	1.0	1.0	869	242	10^4	3	HOT TUBE
DIOH	CsI	1.0	1.34	682	374	10^6	0.4	HOT TUBE
EI	CsI	1.0	1.11	602	292	10^6	0.3	HOT TUBE

[a]Mole fraction of nucleating species at nucleation front.

[b]From pre-test analysis of experiments performed at TREAT reactor (ANL).

[c]Nucleation occurs during mixing with steam.

[d]Aerosol Transport Tests conducted at Marviken facility in Sweden.

[e]Transport tests conducted at ANL.

NOMENCLATURE

A	cross-sectional area
D	diffusion coefficient
G	Gibbs free energy
J	nucleation rate
Kn	Knudsen number
k	Boltzmann constant
k_g	thermal conductivity
L_j	specific (latent) heat of condensation
M	molecular weight
m_j	mass of a condensible molecule
p	pressure
p_s	saturation pressure
R_g	universal gas constant
r	particle radius
S_j	saturation ratio
T_s	particle surface temperature
T_b	gas temperature at the particle boundary layer
T_g	gas temperature
t	time

Greek

α_j	condensation coefficient
β^j	monomer impingement rate
γ	ratio of specific heats
$\delta(r-r^*)$	Dirac delta function
λ	mean free path
ρ	density
σ	surface tension
Φ	energy function

Subscripts

d	deposition
g	gas
j	species
l	liquid

5.3 BEHAVIOUR OF HYGROSCOPIC AEROSOLS

5.3.1 Phenomenology

Introduction

The formation and presence of water soluble compounds will affect the behaviour of fission products in the primary system and in the containment during nuclear power plant core melt accidents. In the primary system a translocation of deposited material may occur due to hygroscopicity, and in the containment steam will condense on these hygroscopic particles even under subsaturated conditions.

Water Activity

The saturation ratio at the particle surface (S_r) can be determined through minimization of the Gibbs free energy. Including the effect of surface tension this leads to:

$$S_r = \frac{p_{v,r}}{p_s(T_r)} = A_w \exp\left(\frac{2\sigma_\ell(T_r)M_w}{rR_g\rho_\ell T_r}\right) \qquad (5.1)$$

Here σ_ℓ denotes the surface tension and ρ_ℓ the density of the liquid droplet, M_w is the molecular weight of water and T_r the temperature at the particle surface. $p_{v,r}$ is the water vapour pressure at the droplet surface and $p_s(T_r)$ is the saturation water vapour pressure corresponding to temperature T_r. A_w is the chemical activity of water, r the particle radius and R_g the universal gas constant.

The chemical activity of water for a solution can be expressed according to the modified Raoult's law:

$$A_{w,i} = \frac{1}{1 + Q_i M_w m_i} \qquad (5.2)$$

where Q_i is the van't Hoff factor. For very dilute solutions Q_i is constant and the saturation ratio can be calculated from equations (5.1) and (5.2) using experimental values for Q_i. m_i is the molality of salt i.

For concentrated solutions one must use experimental data to calculate the water activity, if available. Meissner (MEI80) introduced a correlation to calculate the reduced activity coefficient (Γ) from an extended Debye-Hyckel equation. The water activity (A_w) is then obtained by integrating the Gibbs-Duhem equation:

$$-55.5\ln(A_{w,i}) = \frac{2I_i}{z_1 z_2} + 2\int_1^{\Gamma_i} I_i d\ln\Gamma_i \qquad (5.3)$$

where z_1, z_2 are the charges of the cations and anions, respectively, of electrolyte i. I_i is the ionic strength of a solution. This method gives a fairly good agreement with the experimental data at different electrolyte concentrations and at temperatures from 25 °C to 120 °C. Other methods are more complex and they need at least two adjustable parameters (BAL85).

Several methods in the literature for calculating the water activity of mixed solutions have been compared (SAN74). In this reference a relation developed by Robinson and Bower is recommended, because it gives predictions for the water activity within 1 to 2 %, and is less complicated than other methods.

Mass and Heat Transfer

If simultaneous mass and heat transfer to aerosol particles is considered, one can find the following relations for the droplet temperature and growth rate in the steady state (KRE87):

$$T_r = T_\infty + \frac{LI_T(e^a - 1)}{4\pi r K \beta_T a} \qquad (5.4)$$

$$\frac{dr}{dt} = \frac{I_T}{4\pi\rho_p r^2} = \frac{p_T M_w D\beta_M}{R_g T_\infty r\rho_p} \ln\frac{1 - p_{v,r}/p_T}{1 - p_{v,\infty}/p_T} \qquad (5.5)$$

where I_T is the total mass flow rate directed towards the droplet, L is the specific (latent) heat of condensation of water, K is the thermal conductivity of the gaseous atmosphere and D is the diffusion coefficient of the surrounding gas. Here p_T is the total gas pressure, $p_{v,r}$ is the partial pressure of steam at the droplet surface

and $p_{v,\infty}$ is the partial pressure of steam far from the droplet surface. β_M and β_T are transitional correction factors for the continuum fluxes of mass and heat, respectively. In the case of hygroscopic particles the factor a (a Stefan flow term) is small and may be safely neglected (KRE87). The effect of heat transport by radiation is not taken into account, because of its negligible effect on the growth rate in containment conditions (WAG82, BAR88). Numerical solutions can be obtained by solving eq. (5.4) for T_r and then the droplet radius growth rate (dr/dt) can be calculated from eq. (5.5).

The Clausius-Clapeyron equation can be used to obtain the saturation vapour pressure $p_{s,T}$ when the difference between T_r and T_∞ is less than a few K, and the zeroth-order approximation for the mass flux of eq. (5.5) may be applied when the steam partial pressure is small compared to the total pressure. Thus we get the well known Mason equation for the droplet growth rate (MAS57):

$$r \frac{dr}{dt} = \frac{(S_\infty - S_r)}{N_M/\beta_M + N_T/\beta_T} \quad , \qquad (5.6)$$

where N_M describes the mass transfer effect and N_T the heat transfer effect.

For heat and mass transport the results of eq. (5.6) were compared to eqs. (5.4) and (5.5) at a total pressure of 0.3 MPa and at 99.8 % relative humidity. The growth times for the initially 1.0 µm dry particle were 6 seconds and 100 seconds, respectively. In the NAUA-HYGROS code eq. 5.6 is used.

The Equilibrium Radius

When the behaviour of hygroscopic aerosol particles in a nuclear power plant containment atmosphere is simulated, an equilibrium between the atmospheric R.H. and saturation ratio at the particle surface can be applied under certain conditions.

If there is an insoluble compound in the particle, then the saturation ratio at the particle surface according to eqs. (5.1) and (5.2) is:

$$1 + \left[\frac{2\sigma_1 M_w}{r_e \rho_1 R_g T} \right] 1 + \left[\frac{i M_w f_m \rho_{po} r_o^3}{M_s (r_e^3 \rho_p - r_o^3 \rho_{po})} \right]^{-1} = S_r \quad (5.7)$$

where r_o is the initial radius of the seed particle,

ρ_{po} is the initial density of the seed particle,

ρ_p is actual density of the particle,

m_s is the mass of the soluble component

m_{is} is the mass of the insoluble component, and

$f_m = m_s/(m_s+m_{is})$.

With a little algebra and the use of eq. (5.7) it is possible to find a fourth order equation for the equilibrium radius (r_e): At equilibrium $S_\infty = S_r = S$, $r = r_o$ and $T_r = T_g = T$.

To be accurate one should observe that ρ is a function of r.

$$(S-1) r_e^4 - \left[\frac{2\sigma_1 M_w}{\rho_1 R_g T} \right] r_e^3 + \left[\frac{\rho_{po} r_o^3}{\rho_p} \right] \left[\frac{i S M_w f_m}{M_s} + 1 - S \right] r_e$$

$$+ \frac{2\sigma_i M_w \rho_{po} r_o^3}{R_g T \rho_p \sigma_1} = 0. \quad (5.8)$$

At equilibrium $S_\infty = S_r = S$, $r = r_e$ and $T_\infty = T_r = T$. To be accurate one should observe that ρ is a function of r. An iterative solution for this equation is possible by reproducing the calculation with new values for ρ. For concentrated aqueous solutions i is a function of r and T.

5.3.2 Calculations and Comparisons with Experiments

Introduction

LACE experiments (described in chapter 4) gave valuable information on hygroscopic aerosol behaviour in containment conditions. The models on hygroscopic aerosols in the MAAP and NAUA codes were compared to the results of LACE tests LA2 and LA4. The aim of this comparison was to evaluate whether the simple model used in the MAAP code is suitable for analysing the behaviour of hygroscopic particles in severe accidents, and if not, what modifications are needed to guarantee a reasonable conservatism.

LACE Test LA2

MAAP and NAUA calculations with no steam condensation on particles

LACE LA2 test simulated an accident sequence with pre-existing leakage caused by failure to isolate the containment. A detailed describtion of the actual test conditions can be found in the LACE TR-007 data report (see also Chapter 4).

The data from the LACE report TR-007 were used as input for MAAP 3.0B and NAUA-HYGROS codes to calculate the aerosol behaviour during test LA2 without condensation on particles. This was done to compare a mechanistic aerosol model (NAUA) with the special correlation method used in the MAAP 3.0B code. MAAP does not need the particle source size distribution parameters.

The total suspended mass concentrations calculated by MAAP 3.0B and NAUA-HYGROS are presented in Fig. 5.3.1. It can be seen that the mass concentration predicted by MAAP is lower than that given by NAUA up to 1000 minutes, when the discrepancies disappear. The peak concentration calculated by MAAP is lower by a factor of 1.21 than that given by NAUA, and at 165 minutes this factor is 1.78.

Table 5.3.1 shows the settled, plated, leaked and airborne masses at the end of the LA2 test as calculated by MAAP and NAUA.

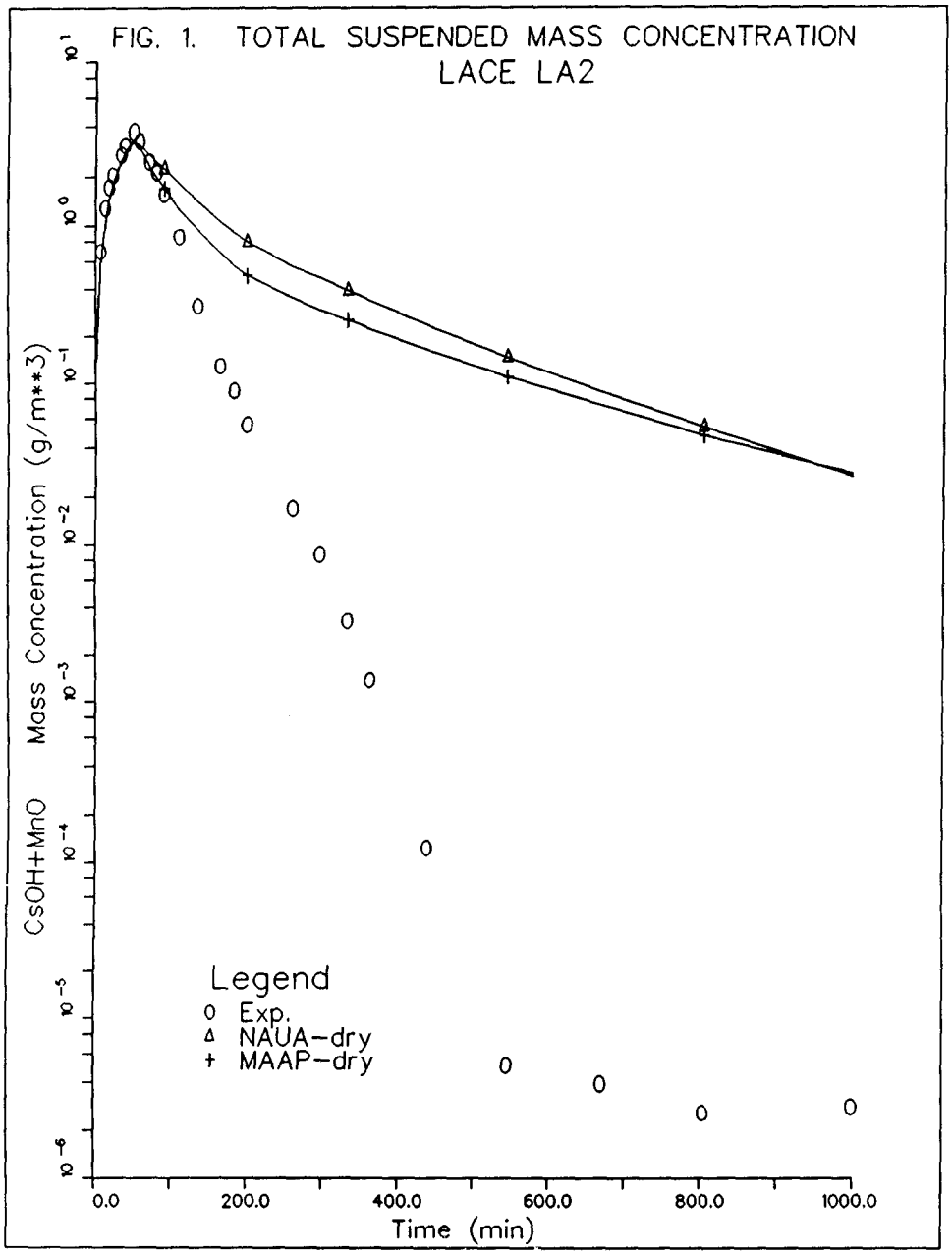
FIG. 1. TOTAL SUSPENDED MASS CONCENTRATION
LACE LA2
CsOH+MnO Mass Concentration (g/m**3)
Time (min)
0.0
200.0
400.0
600.0
800.0
1000.0
Legend
Exp.
NAUA-dry
MAAP-dry

Fig. 5.3.1

Table 5.3.1. Settled, plated, leaked and airborne masses in LA2 test (dry).

Code	Settled (g)	Plated (g)	Leaked (g)	Aerosol (g)	Total (g)
MAAP	1908.0	513.3	1999.9	23.5	3945
NAUA	1599.0	523.1	1793.0	22.5	3938

MAAP and NAUA Calculations with Steam Condensation on Hygroscopic Particles

In the NAUA-HYGROS code the steam condensation rate on hygroscopic particles is calculated according to eq. (5.6).

In the MAAP code the rate of steam condensation on particles is not calculated. Instead the equilibrium radius at a given relative humidity (S_∞) is calculated using eq. (5.8).

In the MAAP model it is assumed that the steam condensed on the particles does not have an effect on S_∞ (ie. $dm_p/dt \equiv 0$). However in the NAUA-HYGROS code the rate of change in the steam mass is taken into account according to eq. (5.6). Thus the change in the steam mass is:

$$dm_s/dt = dm_{in}/dt - dm_{out}/dt - dm_w/dt - dm_p/dt, \quad (5.9)$$

where m_s is the steam mass in the containment, m_{in} is the steam mass source to the containment, m_{out} is the leaked steam mass and m_w is the steam condensed onto containment walls. Even if dm_p/dt is small, it has a dramatic effect on particle size near saturation (ie. $S_\infty \approx 1$).

It is thought that in the LACE tests, the measured steam fraction also includes the water already condensed on particles. This assumption is supported by NAUA-HYGROS (VTT) and St&W NAUA LACE code comparison posttest analyses (Chapter 4). Consequently the measured S_∞ data from TR-007 Table 5.3 were used as input for the MAAP and NAUA-HYGROS codes. However, in the NAUA analysis S_∞ is changed because of the growth according to eq. (5.6).

From Fig.5.3.2 it can be seen that MAAP underestimates the total suspended mass concentration during the first 400 minutes. This is due to overestimation of the saturation ratio S_∞. Table 5.3.2 gives the settled, plated,

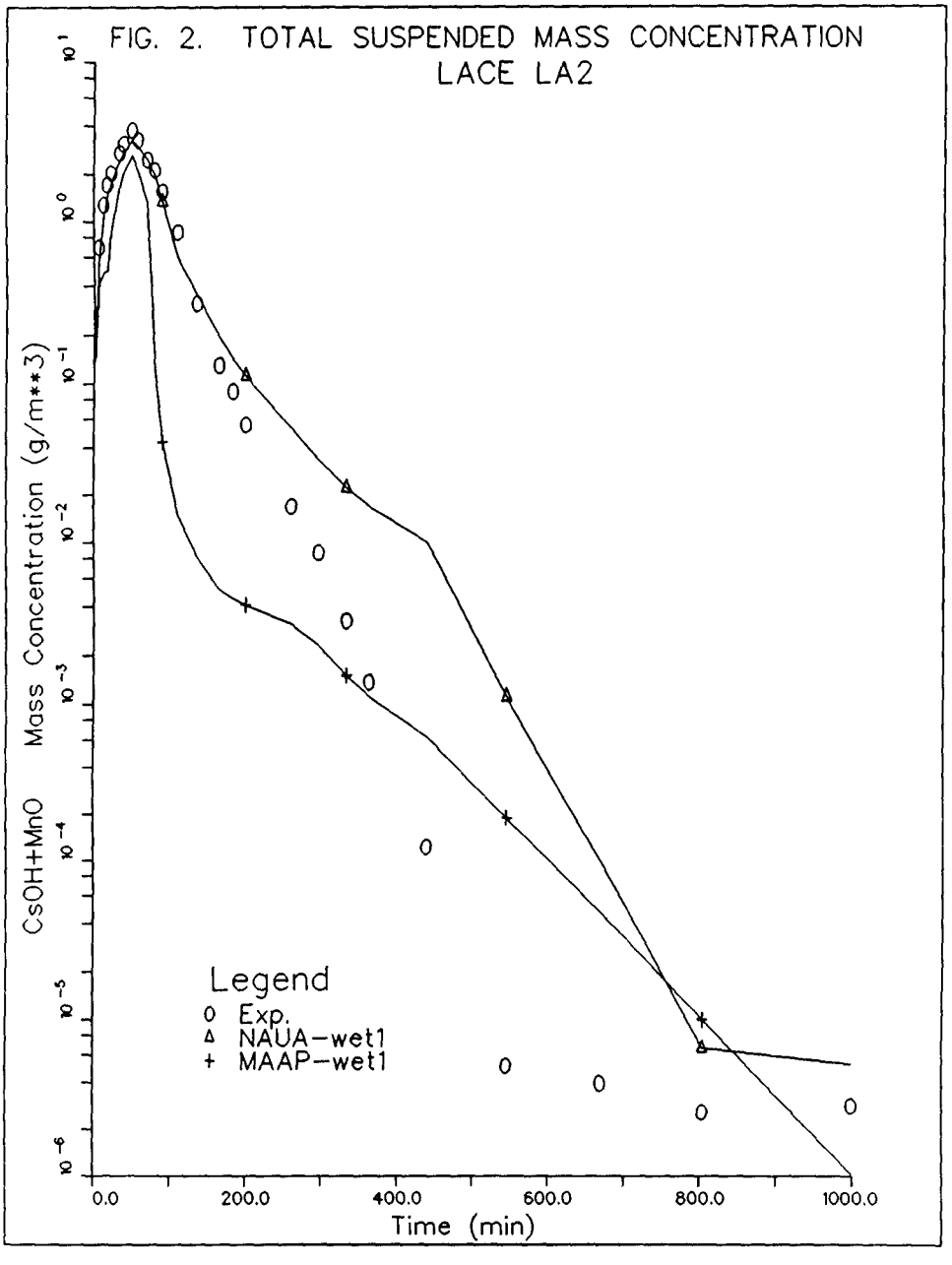
FIG. 2. TOTAL SUSPENDED MASS CONCENTRATION
LACE LA2
CsOH+MnO Mass Concentration (g/m**3)
Time (min)
0.0
200.0
400.0
600.0
800.0
1000.0
Legend
Exp.
NAUA-wet1
MAAP-wet1

Fig. 5.3.2

leaked and airborne masses calculated by MAAP and NAUA at the end of the test.

In the MAAP model, conservative values are used for the van't Hoff factor (≡1) and the initial particle radius (= 0.3 μm). This means that MAAP underestimates the equilibrium radius at certain values of relative humidity. Thus the discrepancies between MAAP, NAUA-HYGROS and the experimental results (Fig. 5.3.2 and Table 5.3.2) are not due to the immediate equilibrium model used in MAAP.

During the analyses of LACE tests it was found that the method of calculating the saturation ratio (S_∞) in the MAAP and NAUA codes is a key factor for understanding the recognized discrepancies, which are mostly due to overestimation of the saturation ratio S_∞ in the MAAP code (Table 5.3.2).

Table 5.3.2 Settled, plated, leaked and airborne masses in LA2 test (wet).

Code	Settled (g)	Plated (g)	Leaked (g)	Aerosol (g)	Total (g)
MAAP	2998.0	340.1	607.4	8.9E-4	3945
NAUA	2271.0	428.8	1238.0	4.4E-3	3938
EXP.	1973.0	449.0	1515.0	2.5E-3	3937

In this analysis it was also recognized that the behaviour of hygroscopic aerosols as calculated by NAUA-HYGROS is very sensitive to the steam input rate, which determines the relative humidity (S_∞) in the containment. From the LA2 data report this steam input rate can be determined in two ways. The first way (NAUA-wet1) is to use the measured steam pressures ($dm_s/dt + dm_p/dt$) from Table 5.3, and the second way (NAUA-wet2) is to use the steam source in the containment (dm_{in}/dt from TR-007 p.51) minus the steam leak rate (dm_{out}/dt from Table 5.8) minus the steam condensation rate on walls (dm_w/dt from Table 5.12). These two ways should be consistent with each other according to eq. (5.9):

$$dm_s/dt + dm_p/dt = dm_{in}/dt - dm_{out}/dt - dm_w/dt,$$

However, these two methods gave totally different steam input rates into the containment vessel. As a result, one can see the effect of relative humidity on the behaviour of hygroscopic aerosols in these two cases (Fig 5.3.3). This demonstrates also how difficult it is to get a correct thermal-hydraulic input for aerosol codes in order to calculate steam condensation on hygroscopic particles.

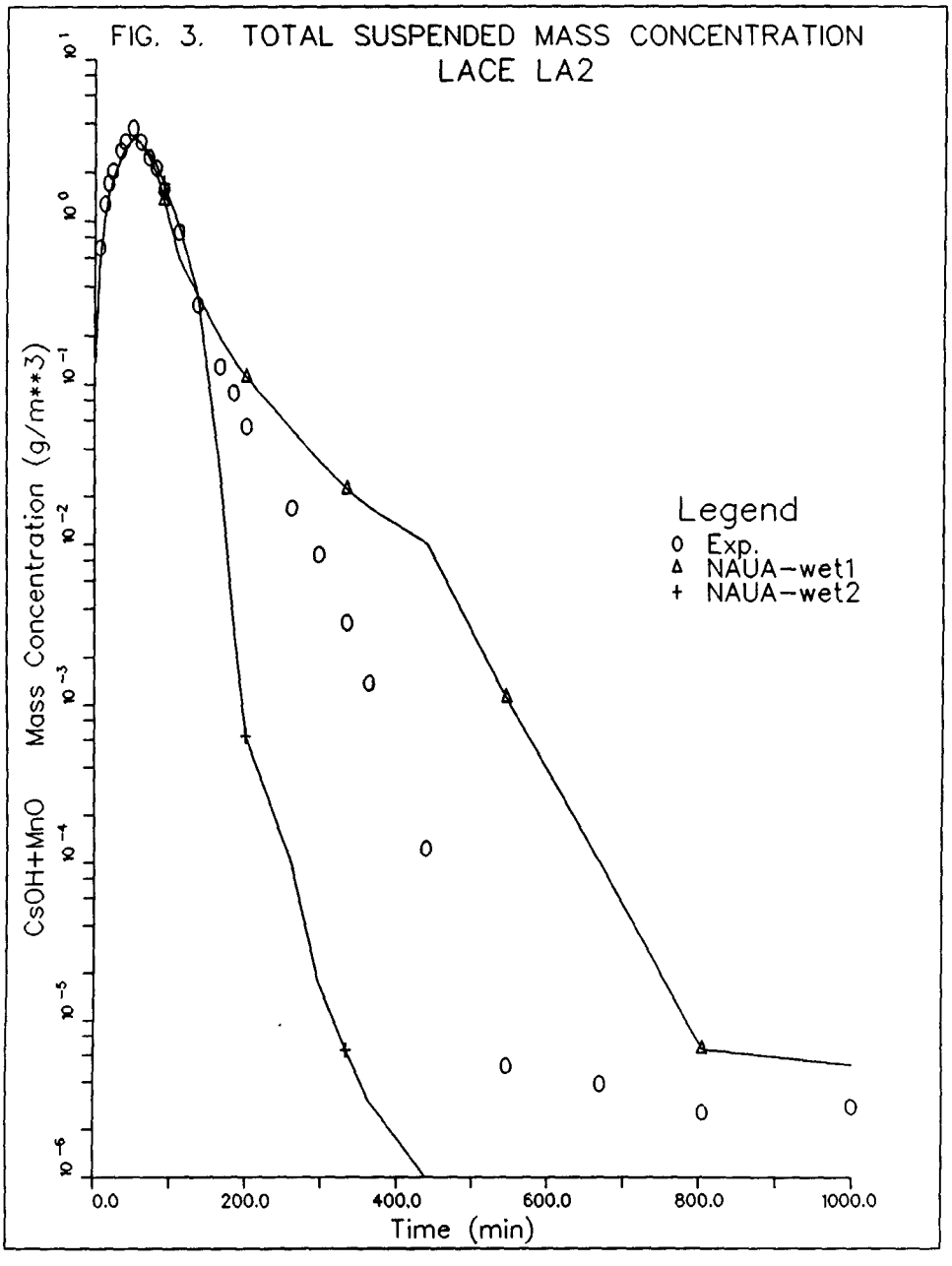
FIG. 3. TOTAL SUSPENDED MASS CONCENTRATION
LACE LA2
CsOH+MnO Mass Concentration (g/m**3)
Time (min)
0.0
200.0
400.0
600.0
800.0
1000.0
Legend
Exp.
NAUA-wet1
NAUA-wet2

Fig. 5.3.3

LACE TEST LA4

MAAP and NAUA calculations with no steam condensation on particles

The LACE LA4 test simulated a late containment failure with overlapping aerosol injection periods. A detailed description of the actual test conditions can be found in the LACE TR-025 data report (see also Chapter 4).

The data from the LACE report TR-025 were used as input for the MAAP 3.0B and NAUA-HYGROS codes to calculate the aerosol behaviour during test LA4 without condensation on particles.

The total suspended mass concentrations calculated by MAAP 3.0B and NAUA-HYGROS are presented in Fig. 5.3.4. It can be seen that the mass concentration predicted by MAAP is lower than that of NAUA after the source is turned off and up to 600 minutes.

After 600 minutes MAAP predicts a higher mass concentration than that obtained with NAUA.

Table 5.3.3 shows the settled, plated, leaked and airborne masses calculated by MAAP and NAUA at the end of test LA4.

Table 5.3.3. Settled, plated, leaked and airborne masses in LA4 test (dry).

Code (dry)	Settled (g)	Plated (g)	Leaked (g)	Aerosol (g)	Total (g)
MAAP	3999.0	845.3	294.8	2.4	5142
NAUA	3558.0	1128.0	447.8	0.9	5135

MAAP and NAUA calculations with steam condensation on hygroscopic particles

In the LA4 analysis the measured S_∞ data from TR-025 Table 6.3 were used as input for the MAAP and NAUA-HYGROS codes, in the same way as in the LA2 analysis.

From Fig. 5.3.5 it can be seen that MAAP underestimates the suspended CsOH mass concentration during and after the injection period. This is due to overestimation of

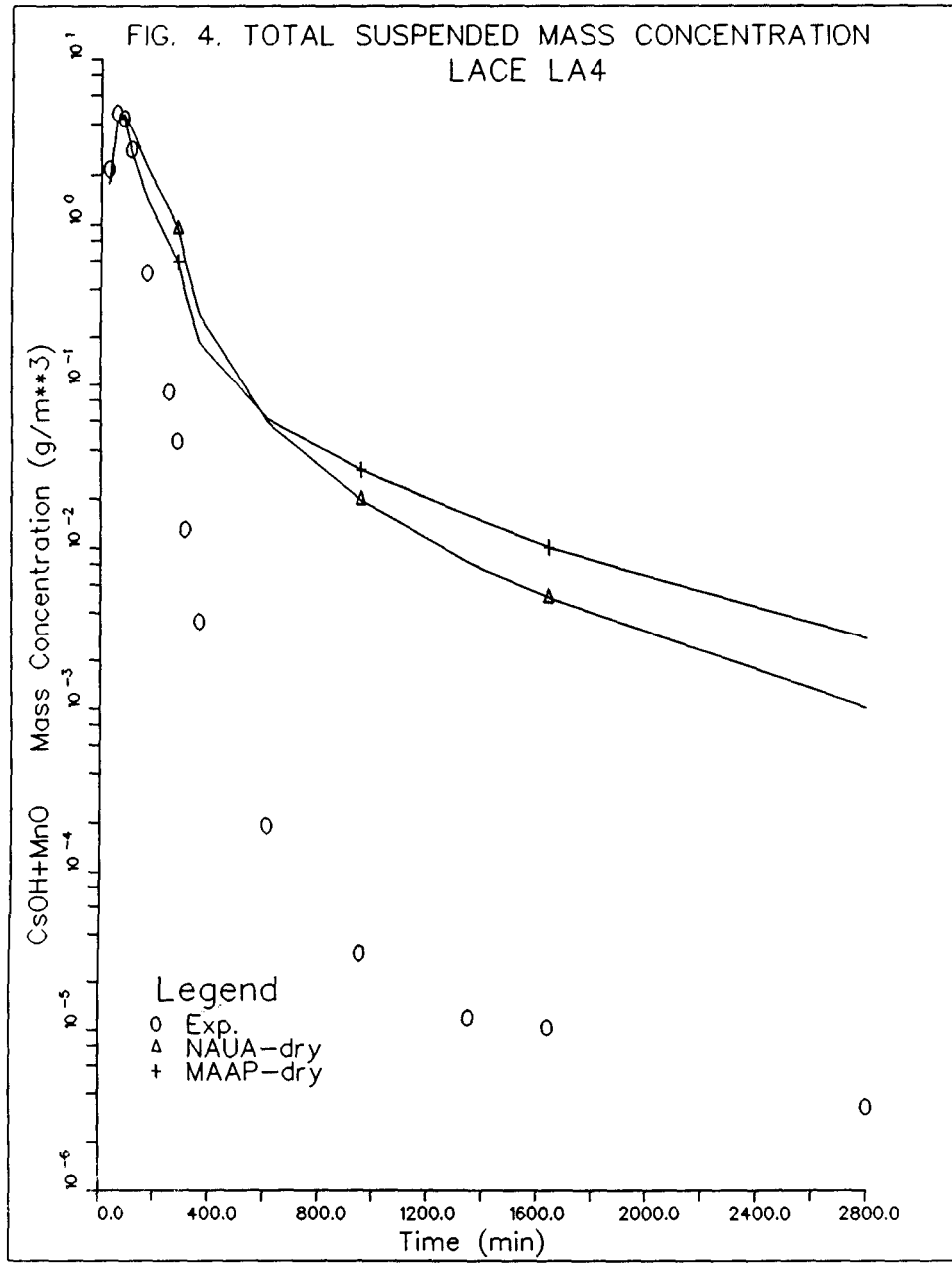
FIG. 4. TOTAL SUSPENDED MASS CONCENTRATION
LACE LA4
CsOH+MnO Mass Concentration (g/m**3)
Time (min)
0.0
400.0
800.0
1200.0
1600.0
2000.0
2400.0
2800.0
Legend
Exp.
NAUA-dry
MAAP-dry

Fig. 5.3.4

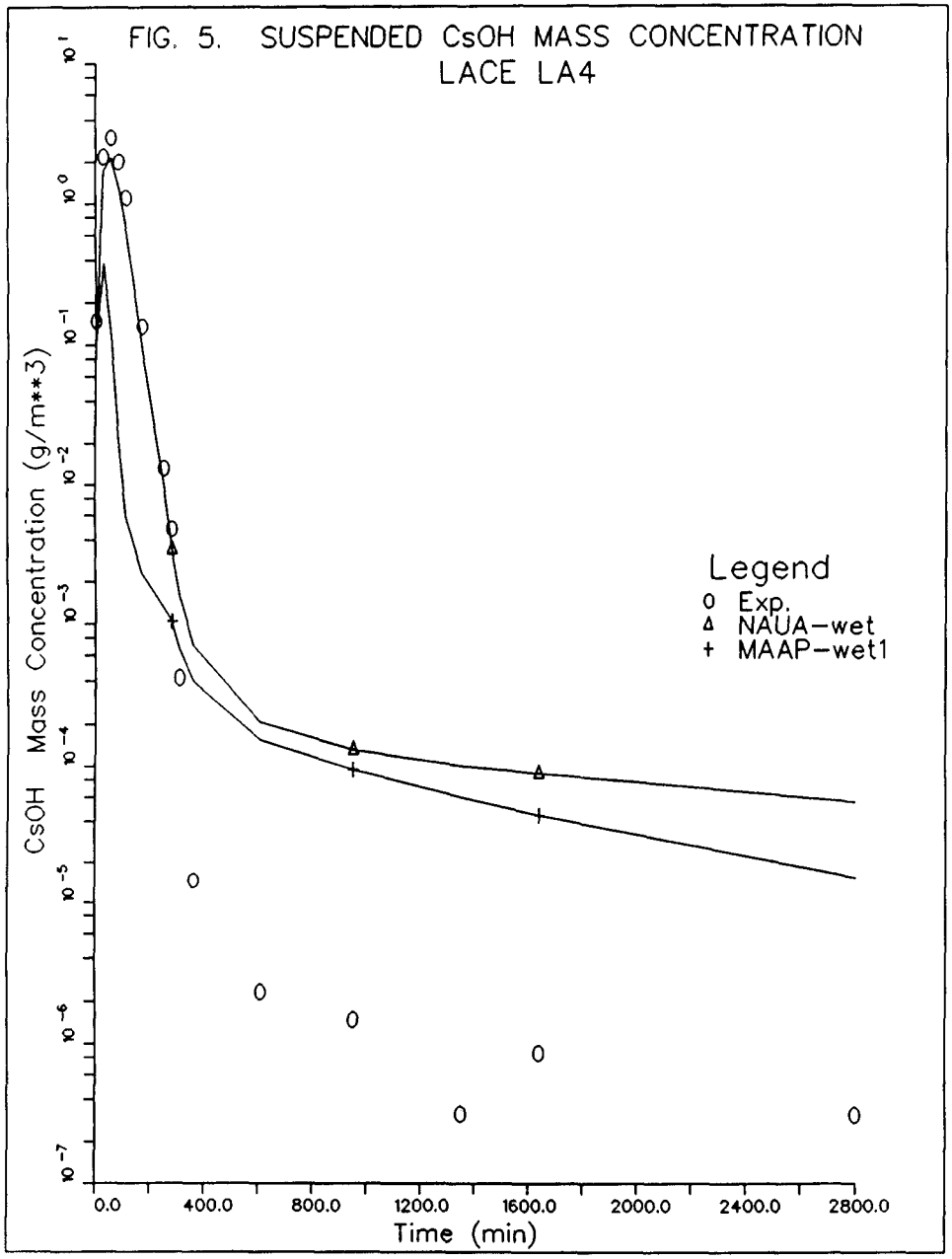
FIG. 5. SUSPENDED CsOH MASS CONCENTRATION
LACE LA4
CsOH Mass Concentration (g/m**3)
Time (min)
0.0
400.0
800.0
1200.0
1600.0
2000.0
2400.0
2800.0
Legend
Exp.
NAUA-wet
MAAP-wet1

Fig. 5.3.5

the saturation ratio S_∞. This means that almost all CsOH is removed before MnO injection starts. Thus at later times there is not much suspended hygroscopic matter (CsOH) and so MAAP starts to overestimate the MnO concentration (Fig. 5.3.6) (in fact it also overestimates the CsOH concentration after venting).

Table 5.3.4 gives the deposited, leaked and airborne CsOH masses and Table 5.3.5 the settled, plated, leaked and airborne CsOH+MnO masses at the end of test LA4.

Table 5.3.4. Deposited, leaked and airborne CsOH in LA4 test (wet).

Code (wet)	Deposited (g)	Leaked (g)	Aerosol (g)	Total (g)
MAAP	2881.0	0.5	5.4E-3	2881
NAUA	2872.4	1.3	4.6E-2	2874
EXP.	2867.0	(a) 4.1	2.6E-4	

Table 5.3.5. Settled, plated, leaked and airborne masses in LA4 test (wet).

Code (wet)	Settled (g)	Plated (g)	Leaked (g)	Aerosol (g)	Total (g)
MAAP	5014.0	110.8	16.7	0.4	5142
NAUA	4599.0	527.5	2.9	0.1	5130
EXP.	4490.0	532.0	(a) 38.5	2.8E-3	

(a) maximum leak calculated from the measured airborne mass at the start of venting (the measured leaked mass also includes the mass deposited along the leak path before venting).

Conclusions and Recommendations

In both the LA2 and the LA4 tests the results calculated with MAAP(dry) were in a reasonable agreement with those predicted by a mechanistic NAUA code (Figs 5.3.1 and 5.3.4). Thus it seems that the MAAP aerosol correlation method can be used for aerosol behaviour analysis in stable containment conditions, when steam condensation onto particles is not taken into account.

When the hygroscopic model is used, MAAP overestimates

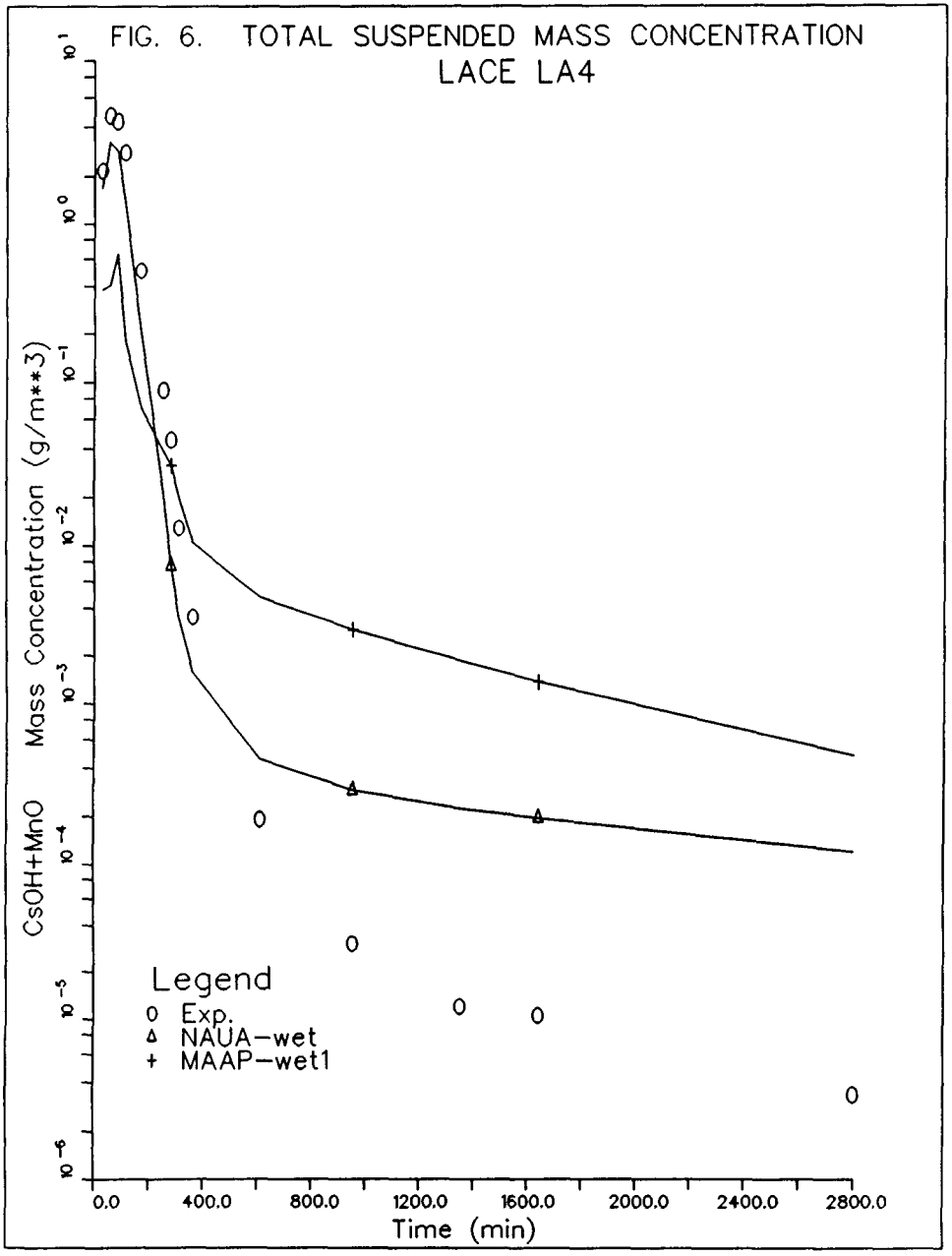
FIG. 6. TOTAL SUSPENDED MASS CONCENTRATION
LACE LA4
CsOH+MnO Mass Concentration (g/m**3)
Time (min)
0.0
400.0
800.0
1200.0
1600.0
2000.0
2400.0
2800.0
Legend
Exp.
NAUA-wet
MAAP-wet1

Fig. 5.3.6

the relative humidity of the containment atmosphere. One reason for this is that MAAP does not calculate the change of relative humidity due to steam condensation on hygroscopic particles. This has a dramatic effect on mass concentration near saturation during the first 400 minutes (Figs. 5.3.2, 5.3.5, 5.3.6). Another reason for the discrepancies between MAAP and NAUA-HYGROS is that according to the mechanistic NAUA code, the particle size distribution is changed after the condensation has taken place and the large particles are deposited. This change in particle size distribution cannot be predicted with the correlations used in MAAP.

During the aerosol injection period hygroscopicity did not seem to have any effect on mass concentration according to the NAUA results, whilst the mass concentration predicted by MAAP changed remarkably when the hygroscopic model was turned on.

On the basis of this comparison, two improvements to the MAAP aerosol model were tried out on an analysis of the LA2 test. First, the change of R.H. due to steam condensation on particles was taken into account, and the hygroscopic model was turned off during the aerosol injection period. Then the minimum time step had to be decreased from 10 seconds to 0.1 seconds in order to get the calculation to converge. This increased the computing time by a factor of 100, but did not eliminate the discrepancies between the experimental and NAUA-HYGROS results (Fig. 5.3.7, MAAP-wet2).

Secondly, the R.H. was limited to a value of 0.99 and the hygroscopic model was turned off during the aerosol injection period. This gave a good agreement with the experimental data during the aerosol injection period, slightly low values for the mass concentration up to 200 minutes and conservative values after 200 minutes (Fig. 5.3.7, MAAP-wet3).

The second modification (max RH=0.99) was also used for the LA4 analysis, and a fairly good agreement with the experimental results was obtained up to 300 minutes; after this MAAP overestimated the mass concentrations (Figs. 5.3.8 and 5.3.9 MAAP-wet3).

In the NAUA-HYGROS analyses of LACE tests it emerged that there was some difficulty in obtaining input data on steam conditions with sufficient accuracy to calculate the steam condensation on hygroscopic particles. The only accepted method of obtaining the steam input data is to use the measured values (LACE data reports). However, these data are inconsistent and different measurements gave different steam input data. (Table IV). This caused considerable changes in the predicted R.H. and mass concentration (Fig. 5.3.3).

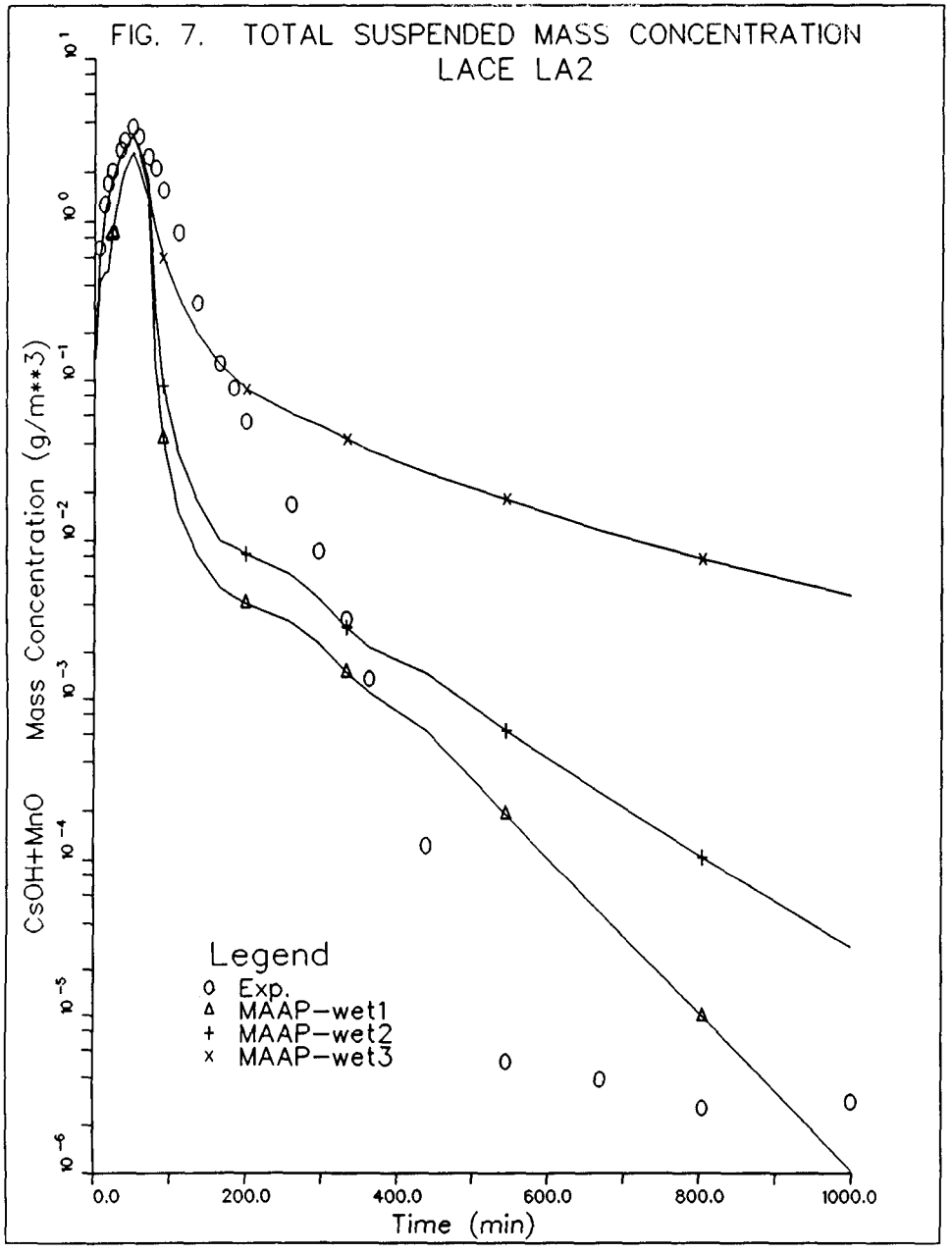
FIG. 7. TOTAL SUSPENDED MASS CONCENTRATION
LACE LA2
CsOH+MnO Mass Concentration (g/m**3)
Time (min)
0.0
200.0
400.0
600.0
800.0
1000.0
Legend
Exp.
MAAP-wet1
MAAP-wet2
MAAP-wet3

Fig. 5.3.7

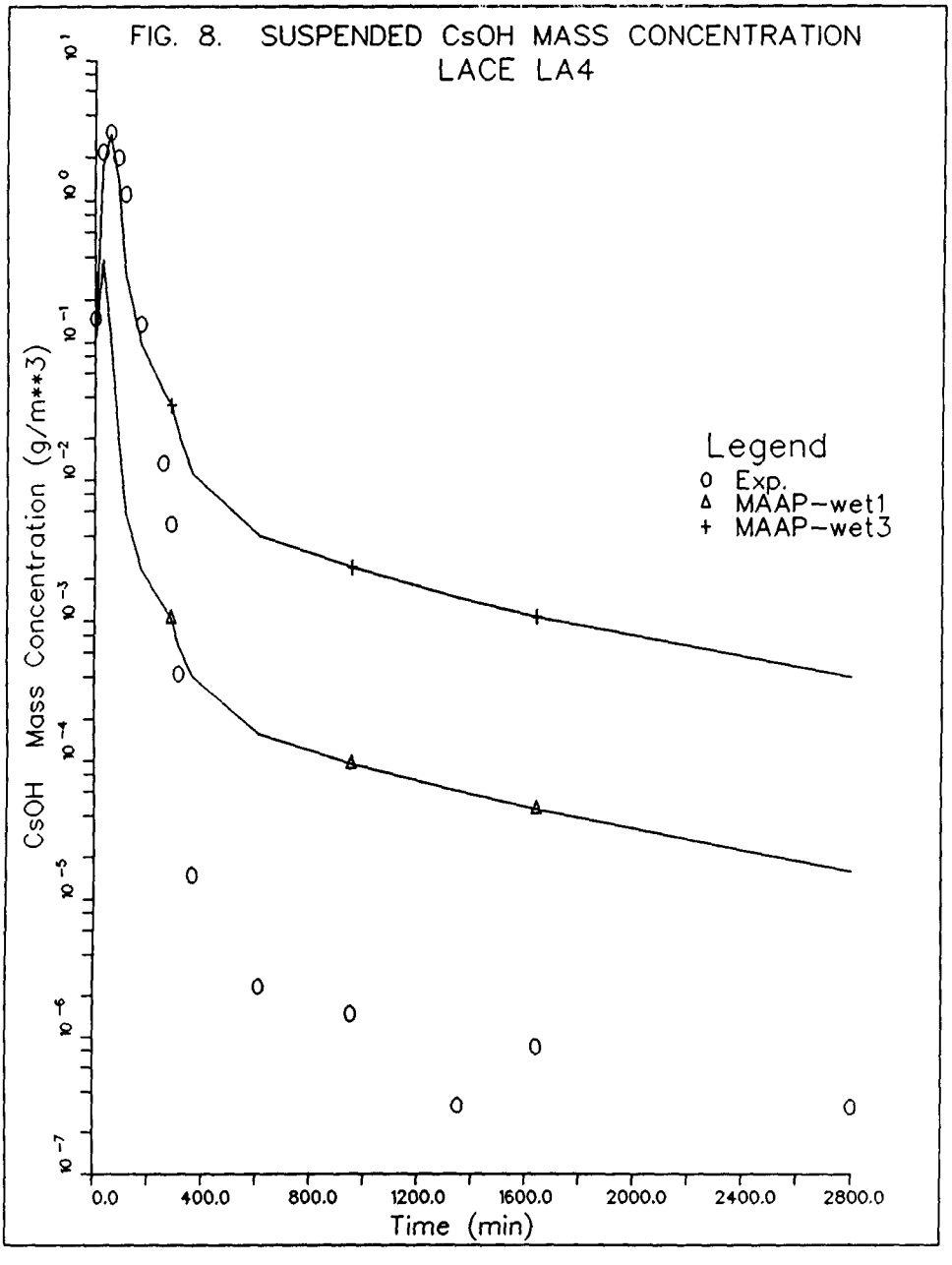
FIG. 8. SUSPENDED CsOH MASS CONCENTRATION
LACE LA4
CsOH Mass Concentration (g/m**3)
Time (min)
0.0
400.0
800.0
1200.0
1600.0
2000.0
2400.0
2800.0
Legend
Exp.
MAAP-wet1
MAAP-wet3

Fig. 5.3.8

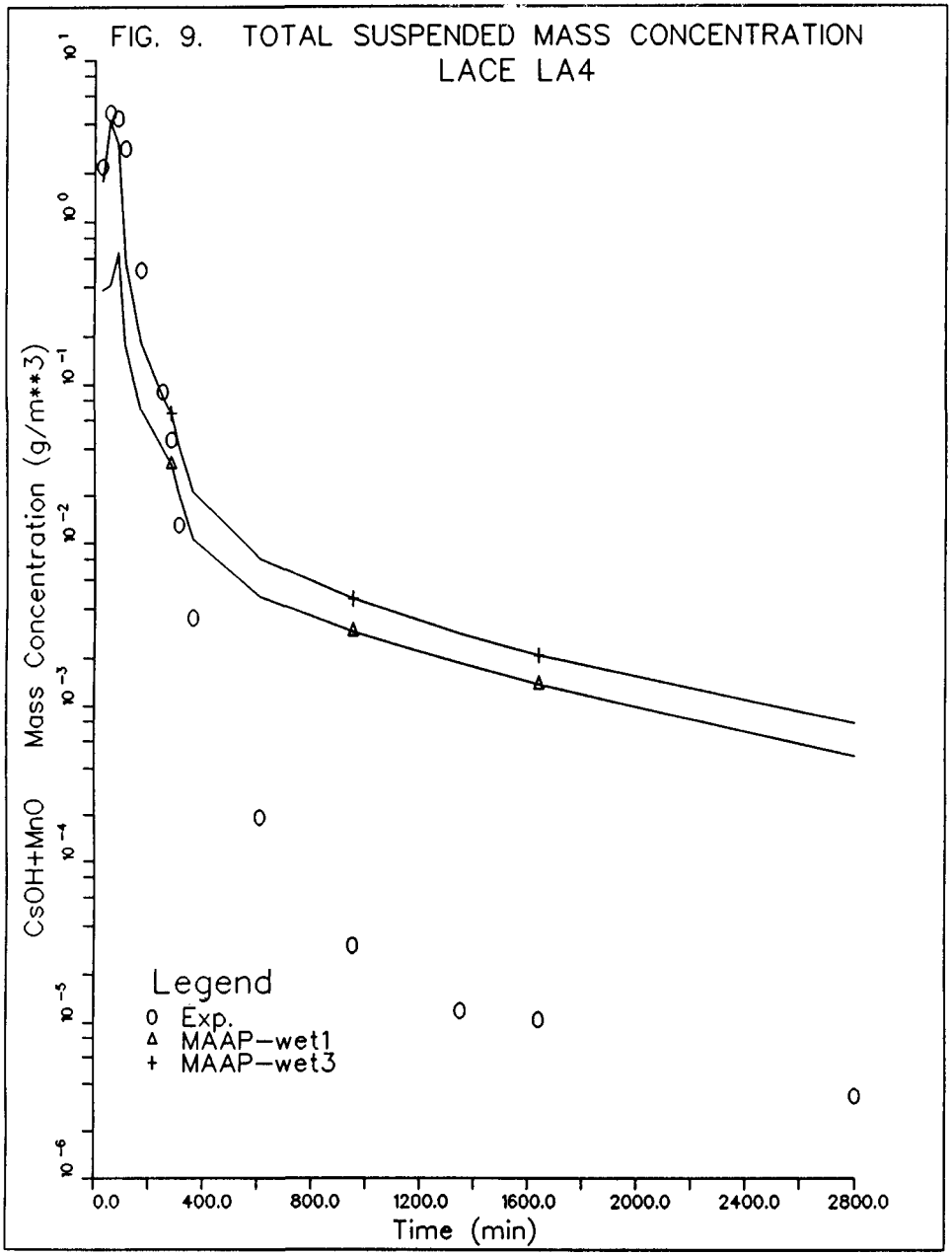
FIG. 9. TOTAL SUSPENDED MASS CONCENTRATION
LACE LA4
CsOH+MnO Mass Concentration (g/m**3)
Time (min)
0.0
400.0
800.0
1200.0
1600.0
2000.0
2400.0
2800.0
Legend
Exp.
MAAP-wet1
MAAP-wet3

Fig. 5.3.9

Because of the recognized sensitivity of hygroscopic models to steam conditions, it is recommended that conservative input parameters should be used (e.g. upper limits for gas temperatures and wall condensation rates) in the analyses of severe accidents.

In the MAAP 3 code it is recommended that the maximum relative humidity should be limited to 99 % and that the hygroscopic model should be turned off during the aerosol injection period.

This comparison between MAAP aerosol correlations and NAUA is valid only for a one compartment case with a constant aerosol source. Preliminary results from the comparison between NAUA and MAAP 3.0B for a Loviisa AB accident sequence indicate that MAAP predicts a much faster aerosol removal rate from the containment atmosphere, by comparison with the results of NAUA.

5.4 POOL SCRUBBING

5.4.1 Phenomenology

It has been well known for a long time that a non-boiling pool traps aerosol particles very effectively, but the decontamination effect in a boiling pool was assumed to be negligible until results were published from experiments at the Battelle Columbus Laboratories (CUN86, MER86, PAU85). These experiments showed that even a boiling water pool can have an appreciable decontamination factor. In connection with the experimental work two computer codes have been developed, SPARC (OWC85) and SUPRA (WAS85a, WAS85b). The MAAP-3 code computes the pool scrubbing effect using tables obtained from SUPRA calculations. At present only SPARC is available for all the countries within the NKA project, and this was made the subject of a special study within group 160. However, the manual (OWC85) is obsolete and therefore a full description of the model could not be made.

The known models for aerosol transport in SPARC are:
- convective flows resulting from the condensation or evaporation of steam
- particle growth caused by water vapour sorption by soluble aerosol material
- sedimentation resulting from gravitational forces
- inertial deposition resulting from centrifugal forces
- diffusional deposition
- mechanical entrainment of pool liquid by the breaking of bubbles at the surface.

The only known thermal-hydraulic model in SPARC is:
- condensation/evaporation of steam based on equilibrium pool temperature.

There is no model for the bubble size or shape; these quantities are input data.

In SUPRA the aerosol models are (WAS85b):
- convective flows resulting from the condensation or evaporation of steam
- particle growth caused by water vapour sorption by soluble aerosol material
- sedimentation
- removal due to flow impingement in the injection zone
- removal due to internal circulation in the injection zone
- inertial deposition in the rising bubble zone
- Brownian diffusion
- thermophoresis
- diffusiophoresis
- desorption from the pool surface.

The thermal-hydraulic models in SUPRA are:
- conservation of mass, vapour species and energy for a binary gas phase
- transfer of gas species through a bubble surface
- pool temporal analysis
- evaporation from the pool surface
- bubble hydrodynamics including shape and volume.

Thus the models in SUPRA are much more comprehensive, especially as regards the thermal-hydraulics.

5.4.2 Calculations

In the test calculations with SPARC the following parameters were varied (HÄG87):

- ratio of major to minor axis of the bubbles
- pool temperature
- ratio of hydrogen to steam flow with constant total gas flow
- value of hydrogen flow when steam flow was constant
- pool depth.

Calculations were also made for comparison with the Battelle experiments (OWC85). Only tin aerosols were considered. The results are given in table 5.4.1. At the end of the project it was possible to make calculations with SUPRA at VTT on two of the tests. The results are given in the table and they show better agreement with the experiment if the humidity is high.

5.4.3 Conclusions

The following main conclusions can be drawn about the SPARC code:

- All the aerosol phenomena which are known to be important are modelled. However, the thermal-hydraulic part is rather primitive, and in particular a model for the bubble shape should be introduced. No chemical or physico-chemical parameters or phenomena are considered. For example, the viscosity of the liquid could be changed both accidentally and methodically, which would influence the bubble sizes. Furthermore, for a full utilization of the code a description of the handling of soluble particles should be added to the User's Manual, which is now obsolete.

- Test calculations have been made with variation of a number of parameters. The decontamination factor was strongly dependent on the pool temperature, the

TABLE 5.4.1

CALCULATIONS ON BATTELLE-COLUMBUS EXPERIMENTS

Exp. No	Steam mass fraction	Measured DF	Calculated DF SPARC	SUPRA
25	0.93	>510	22	127
26	0	110	9.1	11.4
31	0.46	1300	140	
33	0.53	>2100	142	
35	0	3700	298	

noncondensable gas fraction and the depth of the gas injection, while it was nearly independent of the gas flow.

- The calculations made for dry particles indicate that the code gives too small decontamination factors compared to the experiments, but this result is only preliminary, and further investigations should be made. A full evaluation of the results would need a more comprehensive investigation of the code. Some results seem, however, to be in disagreement with SUPRA calculations as well. If it were possible to investigate the SUPRA code, this should give much more insight into the state of the art.

6. ASSESSEMENT OF THE PRESENT STATUS OF KNOWLEDGE

6.1 INPUT DATA

The production of input data for the aerosol codes has been considered by other groups and their accuracy can therefore not be assessed here; however, an estimate will be made of their effect with respect to the required accuracy of aerosol code output data. The desired accuracy of the environmental source term is then set to a factor of 10 if the calculated decontamination factor is 10 000. An estimate shows that this corresponds to an error in the decay constant of 25 %. In addition to the error in the decay constant there are also other sources of uncertainties in the source term. In order to achieve this accuracy, the contribution from input data uncertainties to the error in the decay constant should not be greater than 10 %.

There are four groups of input data for aerosol codes:
* geometrical data
* thermal-hydraulic data
* source data
* chemical data

The uncertainties in geometrical data are mainly concerned with inner surface area, surface roughness and leakage areas. Generally the inner surface area is underestimated and the deposition rate is roughly proportional to the horizontal part of this area. This error may be important in calculating the deposition in the primary circuit and in the lower part of the drywell in a BWR. The surface roughness is normally neglected and this leads to underestimation of turbulent deposition, of importance especially in pipes. Finally, the leakage areas determine the residence time in a given volume and can also determine the possibility for engineered safety measures, for example pool scrubbing. It is therefore important to know their sizes with an error of less than 10 %. Their positions are normally defined by the accident sequence. Even with a careful determination of the geometrical data it is not believed that the corresponding error in deposition rate is within the 10 % range. Large errors in the environmental source term might then arise for late containment breaks.

The most important thermal-hydraulic input data are:

* Total gas flow. If it is given as mass flow it has to be converted to volume flow. In this case the gas density must also be known.

* Steam condensation flow on boundary surfaces and on particles.

* Pressure and temperatures on surfaces and in the gas.

* Gas viscosity. This can also be calculated if the partial gas pressures and the temperature are given.

The total gas flow determines the leakage, and the volume flow should therefore be known with an accuracy of better than 10 %. The accuracy needed for the steam condensation flow is dependent on whether or not particle growth by steam condensation is an important factor for the deposition. It is not believed that this phenomenon can be calculated with the desired accuracy as the saturation ratio is a very uncertain parameter. The pressure and temperature have an effect on the viscosity and gas density, on the thermophoretic effect and on the nucleation process. Only if nucleation is considered do these quantities need to be considered separately, and the accuracy needed is then better than 10 %. The particle transport velocity due to settling and diffusion is inversely proportional to the viscosity. Thus, this parameter should be known within an accuracy of 10 %.

The environmental source term is of course directly proportional to the release rate of fission products from the core debris. As this factor is only one of many uncertain parameters it should be known at least within a factor of 2 for the more important fission products. For aerosol transport calculations the particle size spectrum is also needed. If the source particles are small, there is no high demand on the accuracy of the size spectrum as the particles will agglomerate rapidly. However, if the average source particle diameter is above about 3 μm, the sedimentation will largely compensate for agglomeration and the deposition velocity will be roughly proportional to the square of the source particle diameter. Thus for source particles of this size and larger, the accuracy of the average diameter has to be of the order of 5 %. As this probably cannot be achieved, a lower limit for the actual value should be used. The environmental source term will then be conservative if revaporization is negligible.

The chemical effects on aerosol transport have for a long time been largely neglected. They are, however, important for at least the following mechanisms:

* fission product release from the debris
* hygroscopicity
* chemisorption
* nucleation
* surface transport
* volatility
* heat generation
* solubility in water pools
* resuspension and re-evaporation

No quantitative values regarding their importance for aerosol transport have been assessed, as this area is at a very early stage of investigation.

6.2 State-of-the-Art of Aerosol Physics

The aerosol processes that have most significance for LWR safety are briefly and qualitatively described in Chapter 3. Of these effects, nucleation and condensation (including hygroscopic effects) are considered in more detail in sections 5.2 and 5.3.

The "pure transport mechanisms" are reasonably well understood, although they may be implemented in different ways in the codes. The minor differences include boundary layer thicknesses, the terminal settling velocity for large particles, and the parameter values in the expression for thermophoresis. More serious is the confusion as to which collision efficiency should be applied for gravitational agglomeration. It is now generally accepted that Fuchs's efficiency should not be used for this case, but it still seems to be applied in some codes, and indirectly it may have crept into some of MAAP's correlations, too.

The primary system codes calculate the effects of carrier gas flow and inertia by using accepted models. (However, the treatment of bends and the deposition in turbulent flow have to be improved in most codes.) These effects are rarely considered in containment calculations, partly because they are not thought to be very important and partly because thermal-hydraulic codes for the containment do not supply the necessary information.

It is customary to simply add terms when more than one removal mechanism is active. As shown by Simons and Simpson (SIM88) for the case of diffusion and settling this is only an approximation, albeit not a bad one.

Addition of the agglomeration terms is usually done simply or quadratically (i.e. the square root of the sum of squares). Simons et al. (SIM86) have shown that simple addition is definitely the better of the two for the case of diffusional and gravitational agglomeration. When turbulent agglomeration is included the best procedure may be that proposed by Dunbar and Fermandjian (DUN84), namely to add the turbulent and gravitational terms quadratically and then simply add the diffusion term to the result.

The influence of the ionizing radiation field seems to be considered in only one code, RAFT (IM87). In principle, radiation may influence an aerosol in several ways, for instance by providing nucleation centres, by influencing condensation of polar molecules, and by influencing agglomeration - each of these via ions or electrically charged aerosol particles. However, for the following reasons the effects of the radiation field are expected either to be insignificant or to increase the conservatism of the calculations. (It is assumed that

the aerosol of any macroscopic volume is electrically neutral and that particle charges are too small to induce disintegration by the Rayleigh instability.):

- According to IM87, the estimated ion number density is far too low to dominate homogeneous nucleation.

- Because of the inhomogeneity of the electric field, condensation of polar molecules (such as water) onto charged particles should be affected in the case of small particles, resulting in increased condensation.

- Condensation of ions onto charged particles will depend on the signs of the charges. For an electrically neutral aerosol the net effect should be small, presumably giving an increase in condensation in analogy with the agglomeration case below.

- Condensation of an ion onto a neutral particle will be stronger than the condensation of the corresponding molecule owing to the induced polarisation of the particles and the resulting electric field. Owing to the presence of water all particles can be assumed to be polarisable.

- Agglomeration will be increased. Detailed analysis is difficult, but in the simple cases considered by Lapple (LAP70) the net result is an increase of the agglomeration rate.

In conclusion, a radiation field will generally result in larger particles, which will settle faster, and thus lessen the amount of fission products available for release to the environment.

The codes can generally be divided into two groups with MAAP as a special case. The other codes are then codes for either the reactor coolant system (RCS) or the containment. The weakest points in the RCS codes are the models for transport in pipes, for revaporization and for nucleation. This last mechanism is usually not considered. For the containment codes a model for hygroscopicity is needed, but it is present in only a few codes. In both groups of codes the modelling of effects from circulational flow is rudimentary and should be improved.

6.3 Coupling Between Aerosol Behaviour, Thermal-Hydraulics and Chemistry

In the reactor coolant system the formation of aerosol particles is greatly affected by thermal-hydraulics and chemistry. For example, the gas cooling rate, surface temperatures and chemical form of condensible species will determine aerosol properties and the condensation rate on walls. Thermal-hydraulics (temperature) and aerosol formation (phase changes and deposition) will affect chemical reactions. Fission products and other vapours will affect flow rates and the decay heat will change temperatures.

There are several codes for analysing aerosols and chemistry separately, but only in two codes, RAFT (EPRI) and VICTORIA (NRC) are these phenomena coupled. However, these codes also have limits; RAFT does not have enough chemical species and compounds, and VICTORIA is lacking in the modelling of aerosol formation by nucleation. Lately these codes have been coupled with thermal-hydraulic and fission product release models. RAFT is coupled with PSAAC and CORMLT codes and is now called SIAM. NRC has coupled VICTORIA with TRAC and MELPROG codes.

In the containment the main interaction between chemistry and aerosols is related to the behaviour of hygroscopic particles, and gaseous iodine interaction with aerosols and walls. From the point of view of thermal-hydraulics the interaction with aerosols is usually not important. However, minor changes in e.g. gas temperatures due to steam condensation on particles can have a dramatic effect on aerosol behaviour.

The correct modelling of steam condensation onto aerosol particles means that the mass and heat transfer of water vapour onto particles and onto containment walls have to be solved simultaneously. This analysis should include the correct treatment of hygroscopic and non-hygroscopic matter.

So far, the heat and mass transport on particles is only modelled correctly in the NAUA-HYGROS (VTT/EPRI) and NAUA (ST&W) codes (these models correspond to the state-of-the-art of aerosol physics). There are no codes where aerosol behaviour and thermal-hydraulics are fully coupled. However, in future CONTAIN will be a code with more or less complete coupling.

MAAP has a certain coupling between thermal-hydraulics and aerosols, to the extent that the fission product heating is considered. This affects all temperatures but can also cause revaporization of the fission products. Because it can occur late in the accident sequence re-vaporization is important, even if the vaporized mass is

rather small.

In the case of aerosol transport several compartments are considered as one node if they are strongly coupled through circulational flows. The release of latent heat by water condensation on particles is not considered.

6.4 Engineered safety features: pool scrubbing, spray, filtered venting, ice condensers

Within the AKTI-160 project the investigation of pool scrubbing was mainly concentrated on the SPARC code, sponsored by the US-NRC (OWC85). Test calculations were done for comparison with experiments made at the BMI, Columbus (PAU85). These calculations indicated that the code underestimated the decontamination factor by about a factor of 10. The SUPRA code, developed for EPRI (WAS85b) has a more comprehensive model for thermal-hydraulics and it seems to give better results when the gas humidity is high. Data from calculations with this code are used in tabular form in MAAP-3, but verification of the pool scrubbing model in the Nordic versions of MAAP-3 has not been done.

Water spray is a very effective engineered safeguard system and was investigated in the CSE experiment (AEC75). A model based on these experiments is contained in the CORRAL code. MAAP-3 contains a simplified model, and the accuracy of this is not known for the AKTI-160 group.

The effectiveness of filtered venting is very dependent on the system. For the Barsebäck FILTRA it is believed that the uncertainty in the estimation of filter efficiency is a factor of 100 in the positive direction and a factor of 10 in the negative direction. This is based on the fact that the effect of steam condensation was not considered. No estimations have been made for other systems.

A study on the retention of iodine and particles within the ice compartment of an ice condenser containment has been made by Winegardner et al. (WIN83). This study was purely theoretical and no experimental studies or more recent theoretical ones are known. Thus, it might be hazardous to assume any credit for ice compartment retention. However, promising experimental results from Battelle are expected.

7. CONCLUSIONS

This report presents conclusions from work in the following areas of aerosol transport behaviour:

- Experiments and predictions of experimental results. Four experiments are considered, namely CSE, Marviken-V, DEMONA and LACE.
- Theoretical and computational studies
- Nucleation and condensation, including behaviour of hygroscopic aerosols
- Pool scrubbing
- Assessment of the present status of knowledge in this field.

The codes considered in the comparative investigations were NAUA-4 and NAUA-5 (discrete particle size distribution), HAARM and RETAIN (log-normal distribution) and MAAP (correlational models for steady state and ageing aerosols). The purpose was to obtain examples of differences in the results produced by codes using different representations of the aerosol size distribution. The main conclusions were:

* Log-normal models may be acceptable for use when leakage and/or source input is the dominating mechanism.
* Log-normal models predict too low releases compared to codes with discrete size classes if containment leaks occur late in the accident. In reality the error may go in both directions, depending on, for example, the hygroscopicity effect.
* The results obtained with MAAP were in reasonable agreement with those from NAUA if dry aerosol and a constant source rate was assumed. Discrepancies were obtained for a varying source rate, particularly in multicompartment cases. The results with high humidity and a hygroscopic aerosol are erroneous by some orders of magnitude.

In the case of aerosol nucleation, some theoretical investigations have been made and the models in the RAFT code have been studied. The nucleation process seems to be important in the primary circuit and the resulting aerosol source has a particle size spectrum which is strongly dependent on the temperature gradient, the pressure and the aerosol concentration. Mass median diameters can vary between 0.1 and 30 μm or more.

The main work on the hygroscopicity effect has been done in another connection, but the work has been followed up during the meetings. It has been concluded that the

NAUA-HYGROS code represents an essential progress in achieving agreeement between experiments and calculations on the aerosol behaviour in a containment. However, good accuracy can only be obtained if the thermal-hydraulic conditions are well known.

Pool scrubbing was studied mainly by making some test calculations with the US-NRC computer code SPARC. The resulting decontamination factors were generally about one order of magnitude smaller than experimental values. It was also recognized that the EPRI code SUPRA has a more comprehensive model for the thermal-hydraulic behaviour in the pool and it seems to give better results when the gas humidity is high.

An assessment of the present status of knowledge has been made and it was concluded that the pure aerosol transport mechanisms are quite well known and are considered properly in a number of computer codes. However, the coupling between aerosol behaviour, thermal-hydraulics and chemistry is essential and such models are still in a preliminary stage. Furthermore, pure aerosol codes need thermal-hydraulic input data with good accuracy. It is therefore not believed that the external source term can be calculated within an accuracy of one order of magnitude if the decontamination factor is 1000 or more. In particular, the tendency to underestimate the decontamination factor is probably high. However, some very unknown mechanisms, as for example re-evaporation and resuspension, may lead to overestimation of the decontamination factor.

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