THE ROLE OF AEROSOL BEHAVIOR IN LWR-CORE-MELT ACCIDENTS

by

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I. Introduction

Recently, a rapidly increasing interest has been observed in nuclear-reactor accident sequences which were - and still are - labeled 'hypothetical'. Regulatory and other authorities have been alerted and are now demanding more detailed and realistic information on core melt sequences and the associated risk. This evolution creates a new situation for that part of reactor safety research which has been directed towards the better understanding of core melt accidents: The application aspect becomes more important.

The transport and the behavior of fission products play a central role in risk evaluation for core-melt-accidents (CMA). In CMA's the amount of released fission products may be so high that a coarse conservative description of fission product behavior leads to very high environmental risks. Therefore, credit has to be given to those natural processes removing airborne fission products from the containment atmosphere. Models for fission product behavior together with models for fission product release and containment response will become increasingly used for better description of CMA sequences and consequences. Also, a better understanding of fission product behavior phenomena may lead to possible countermeasures for mitigating the consequences of such accidents.

With respect to their natural behavior fission products can be classified in gaseous and condensed (solid or liquid) elements or compounds (aerosols). The behavior of nuclear aerosols composed mostly of solid fission products and usually larger amounts of non-radioactive material differs completely from that of the fission gases. Also the radiological impact is different because aerosol particles, once they have been plated out in the surroundings of a power plant, will stay there and contribute to the radiological impact for longer times than the short term effects of noble gases and iodine.

In this paper recent results on the behavior of nuclear aerosols in the post-core-melt accident atmosphere of a PWR-containment are discussed. They represent intermediate information of a long-term program (NAUA-program) carried out in the Laboratory of Aerosol Physics and Filter Technology of the Nuclear Research Center Karlsruhe to understand and describe natural removal processes of nuclear aerosols in condensing steam atmospheres.

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II. Aerosol processes in post-accident containment atmospheres

Airborne particulate material (aerosols) in confined systems undergo a number of interaction and removal processes depending on their number concentrations, particle sizes, external forces and the presence of condensable vapors. Of special significance are the growth of particles by agglomeration and condensation since particle size is a critically important factor in particle behavior. In particular, the following aerosol processes take place:

- agglomeration or coagulation (Brownian, gravitational)
- condensation
- evaporation

leading to change in particle size and

- diffusion
- gravitational sedimentation
- thermophoretic deposition
- inertial/turbulent deposition

leading to removal of particles from the contained atmosphere. Other aerosol processes are considered to be of minor importance in the case of nuclear aerosol behavior [1, 2], although some processes (e.g. resuspension, diffusiophoretic deposition) need further investigation.
Not all these processes do occur in a post-accident LWR containment at all times. Coagulation takes place only at higher particle number concentrations. Condensation of vapors or steam onto particles or containment walls depends on the thermodynamic conditions in the carrier gas (supersaturation). Evaporation of liquid particles needs sufficiently high temperatures. Removal processes depend on temperature distribution and gradients near the walls, flow conditions and containment geometry as well. Of course, several physical properties of the particulate material, of the carrier gas and of existing vapors determine to what extent which process is involved.

Furthermore, all aerosol processes have been treated mathematically so far by assuming spherical particles only. This requires the use of correction factors (shape factors) for real nuclear aerosols, since most of them - except droplets - have irregular shapes.

In summary, nuclear aerosols in the post-accident atmosphere of a LWR represent a three-phase, highly dynamic system composed of solid particles, droplets, vapor and carrier gas (air) which changes rapidly its properties by natural aerosol processes, external forces and geometrical random conditions. In describing such a system with time not only the aerosol processes have to be taken into account but also several process coefficients, temperature time function and related thermodynamic parameters.

In the ne chapter a computer model is described which has been developed in the recent years by taking into account all relevant aerosol processes, thermodynamic parameters and influence factors of core-melt accident scenarios.

III. The NAUA-Model

The purpose of the NAUA-code is to describe the behavior of aerosols under the conditions of a CMA. Since the scenarios of such an accident can be very different, the code has to be able to calculate the aerosol behavior under all these circumstances. Therefore an only empirical model cannot be used since calculations and experiments have shown that the response of an aerosol system on different boundary conditions is quite complicated. Especially it is impossible to assume any simple relation between the source term and the aerosol mass concentration as a function of the time. The decay of the mass concentration influences the mass being able to leak out of the containment and the mass and the related radioactivity released at the time of the containment failure due to overpressurization. The NAUA-code therefore treats as far as possible all physical processes recognized as important simultaneously and is by this reason able to calculate all different accident scenarios which are only input for the code.
The processes being of importance under the conditions of a CMA were already listed in the previous chapter. They are the coagulation, the condensation of water vapour onto the particles, the sedimentation and less important the diffusion and the thermophoresis because of the lack of great temperature gradients in the containment.
The simultaneous action of all these processes under the assumption of homogeneous mixing of the atmosphere can be described by a integro-differential equation [3]. This integro-differential equation can be transformed into a system of coupled first order differential equations if the particle size distribution is appoximated by a number of monodisperse fractions. By this method it is possible to solve the equation numerically stable and to handle arbitrary mathematical expressions for every single process as well as arbitrary input data for the aerosol source and the thermodynamic conditions. Concerning the aerosol source all possible size distributions, lognormal, Gaussian, monodisperse or just arbitrary tables, and every time function for the release can be handled without any restrictions. Since the aerosol particles, being expected during the CMA, are composed of a solid fraction (fuel, steel, concrete and fission products) and a liquid fraction (water), the code takes into account a size dependent composition of the particles. This means that the contents of water and solid material are averaged only over the size of each size fraction but not over the whole size distribution. This procedure allows the computation of condensation as well as evaporation of water onto or from the particles respectively. The radioactive non-volatile nuclides are assumed to be homogeneously distributed over the solid fraction of the particles. This should be justified by the high coagulation rate taking place due to the high particle number concentration. The possible reaction of the volatile fission products (J, Cs) with the particles and droplets are at present not taken into account. A schematic diagram of the code is shown in Fig. 1.

The greatest problem is the exact treatment of the steam behavior and the corresponding influence on the particles. The calculation of steam condensation onto particles under itself welldefined thermodynamic conditions is a solved problem and is experimentally verified [4]. However, these conditions are not

well defined enough for the CMA because most containment codes assume a steam saturation equal or less than 1. This procedure excludes the supersaturation, the NAUA-code needs to calculate the condensation onto the particles. Therefore for code calculations presented in this paper a simple method was used to handle steam sources as well as the temperature function [Fig. 2] given by the containment code [5]. This procedure calculates the condensation onto the particles due to the supersaturation by adding the steam isothermally to the containment atmosphere. This condensation however leads to a temperature increase due to the release of latent heat and reduces the saturation. This saturation is held constant and the temperature is adjusted to the temperature given by the temperature function. The excessive amount of steam is assumed to be lost, by condensation on the walls.
The effect of the condensation on the decay of aerosol mass concentration is dependent on the scenario especially on the times when particles and steam are released.

IV. Results obtained by NAUA calculations for reference hypothetical core-melt accidents in PWR's

The condensation of steam onto particles has been experimentally investigated, condensation rates and efficiencies have been measured for a large band of thermodynamic conditions using different aerosols (including UO_2). The experimental data were in excellent agreement with our calculations in all cases [4]. Therefore, the microphysical description of the condensation process in the NAUA model is considered to be correct. The remaining uncertainties are caused without exception by the insufficient accuracy of the available data on thermodynamic boundary conditions as temperature, steam content or local steam distribution inhomogeneities. We will discuss this further below.

Encouraged by the results of the experiments we performed a series of calculations for a typical core melt accident. As a reference scenario we chose a melt down accident in a 1300 MWe PWR (Biblis B). The accident starts with a double ended break in one primary coolant pipe. After blowdown the ECCS initially operates but fails when switching to the sump. This event is time zero for our calculations because no significant amount of aerosols is produced earlier. The sequence of aerosol and steam releases is shown in Fig. 3. During the evaporation of the water in the reactor pressure vessel (RPV) the core starts to melt and an increasing release of aerosols is assumed. The total mass of released aerosols from this phase is assumed to be 800 kg.

The next phase is characterized by a very low aerosol release, set equal to zero in our calculations. This is when the RPV is boiled dry but has not yet failed. Aerosols are generated but, due to the lack of steam flow as a vehicle, are not transported into the containment.

When the RPV finally fails at 7000 seconds steam from the core concrete interaction sweeps the aerosol content of the RPV into the containment. Additionally fuel and concrete aerosols are produced for a short time as long as the temperature of the melt is sufficiently high. In this phase

200 kg of fuel aerosol and 110 kg of concrete aerosol are produced. It has to be emphasized that these mass figures represent the total suspended aerosol masses of which only 5.97 % are solid fission products. The rest is inactive fuel, cladding and structure material.

Also indicated in Fig. 3 are the values of the mean radius r_g and variance σ of the particle size distribution of the aerosol source term. These values are only rough estimates. However, we have been able to demonstrate that for our reference scenario the particle size and size distribution of the aerosol source has no influence on the overall aerosol behavior for mean particle sizes below $\sim$ 3 µm [6].
Aerosol behavior is much more influenced by the total released mass: higher initial mass concentrations decrease faster with time than smaller initial mass concentrations [1, 2, 7]. Because accurate release data are not yet available the total aerosol mass was parameterized, assuming 2000 kg, 1000 kg and 100 kg total mass (fuel only).
The steam source was assumed to be constant at a rate of 15 kg/sec from 0 sec to 7000 sec. The method of calculating condensation phenomena due to a steam source was explained in chapter III. Keeping in mind that data on temperature and steam concentration in the containment atmosphere are insufficiently known the use of a constant steam source function does not worsen the results further. In fact the sensitivity of calculated results to those assumptions which influence the steam condensation is the greatest. This is easily explained by the results in Table I.

Table I gives the integral leaked aerosol mass from the containment caused by a constant leak rate of 0.25 Vol %/day.
The calculations were performed for the above explained three values of the total released aerosol mass, given in the first column of the table. The integral leaked masses for the reference cases, i.e. with the temperature function in Fig. 2 and the constant steam source of Fig. 3, are listed in the third column. 1146 g escape through the leak when 2000 kg were released, 711 and 156 g with a release of 1000 kg and 100 kg resp. It is an important result that the released masses and the integral leaked masses are not proportional. Comparing e.g. the 2000 kg and 100 kg cases, an increase in released mass by a factor of 20 leads to an increase in leaked mass by a factor of $\sim$ 7 only. This reduction is due to natural aerosol behavior and indicates the benefit of using complex mechanistic codes.

The second column of Table I gives the leaked mass calculated for the case that no steam source is present at all times. These cases without condensation effects are nevertheless interesting because they represent the upper limit of all possible leaked mass values. Whenever condensation on aerosols takes place it leads to a reduction in airborne mass and consequently in leaked mass.

The comparison of column 2 and 3 indeed shows higher values in column 2 in all three cases, the differences being small, however. The reason for the small differences is twofold: Firstly, the steam source acts only two hours, a short time compared to the periods of interest. Secondly, the knowledge of containment temperature and steam saturation functions (input for the NAUA code) is limited, as explained above. Here, the results of our current measurements of steam transport data will certainly improve the results of the calculations.

The last column of the table represents a quite different situation. Here*) no steam source was assumed but instead a decreasing temperature function for the containment atmosphere was used. The function was taken from [8] just as an example without considering its applicability to our case. This forced temperature decrease might be due to containment sprays. With such a forced temperature decrease the NAUA model, of course, calculates a strong condensation and an enhanced aerosol decay. The leaked masses are then lower by a factor of $\sim$ 3.

Table I gives only the leaked mass integrated over all times. Of some interest is also the time dependence of the leaked mass. This is shown in Fig. 4 which represents the 1000 kg case in the second column of Table I, i.e. without steam source. It is important to note that the final value of the leaked mass is reached almost after half a day, and that later on only insignificant amounts are added to the leaked mass. The reason is clearly the strong decay of the airborne mass in the containment which is also shown in Fig. 4. Since the rate of leaking mass is proportional to the airborne mass it will decrease with decreasing aerosol inventory of the containment.

It is further important to emphasize that the decay of the airborne aerosol mass in Fig. 4 is due to natural processes only. The action of appropriate mitigation features might reduce the leaked masses further. However, Fig. 4 shows clearly that such engineered features must operate early. If they start later than 12 hours after the start of the accident they will reduce only the airborne mass but not the leaked mass.

V. Conclusions

The NAUA code describing nuclear aerosol behavior in LWR post-accident containment atmospheres as function of aerosol, thermodynamic and other parameters became recently fully operable. The essential physics in the code has been experimentally verified.

- The NAUA code is based on first principle aerosol physics without any restrictions. All realistic or hypothetical accident scenarios in dry or steam filled containments can be handled by the code provided the thermodynamic parameters are known.

- In calculating various core-melt accident scenarios it became clear that of major influence to the leaked aerosol mass are
  - the released mass from core
  - the steam source and the temperature function in the containment

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*) $T(t) = 54 + 75 \cdot \exp(-4t)$; T [°C], t [hours], (dashed line in Fig. 2)

- In this paper a LOCA with subsequent CMA was considered with the following results:
  - there is no proportionality between released mass from core to leaked aerosol mass
  - main aerosol leakage occurs in the early times of the accident
  - the leaked aerosol mass is reduced by the effect of condensing steam

- To obtain more accurate quantitative results on activity release to the environment (radiological source term) due to aerosol leakage in core-melt accidents a more accurate description of the thermodynamic boundary conditions, in particular in the early phase of the accident, is needed.

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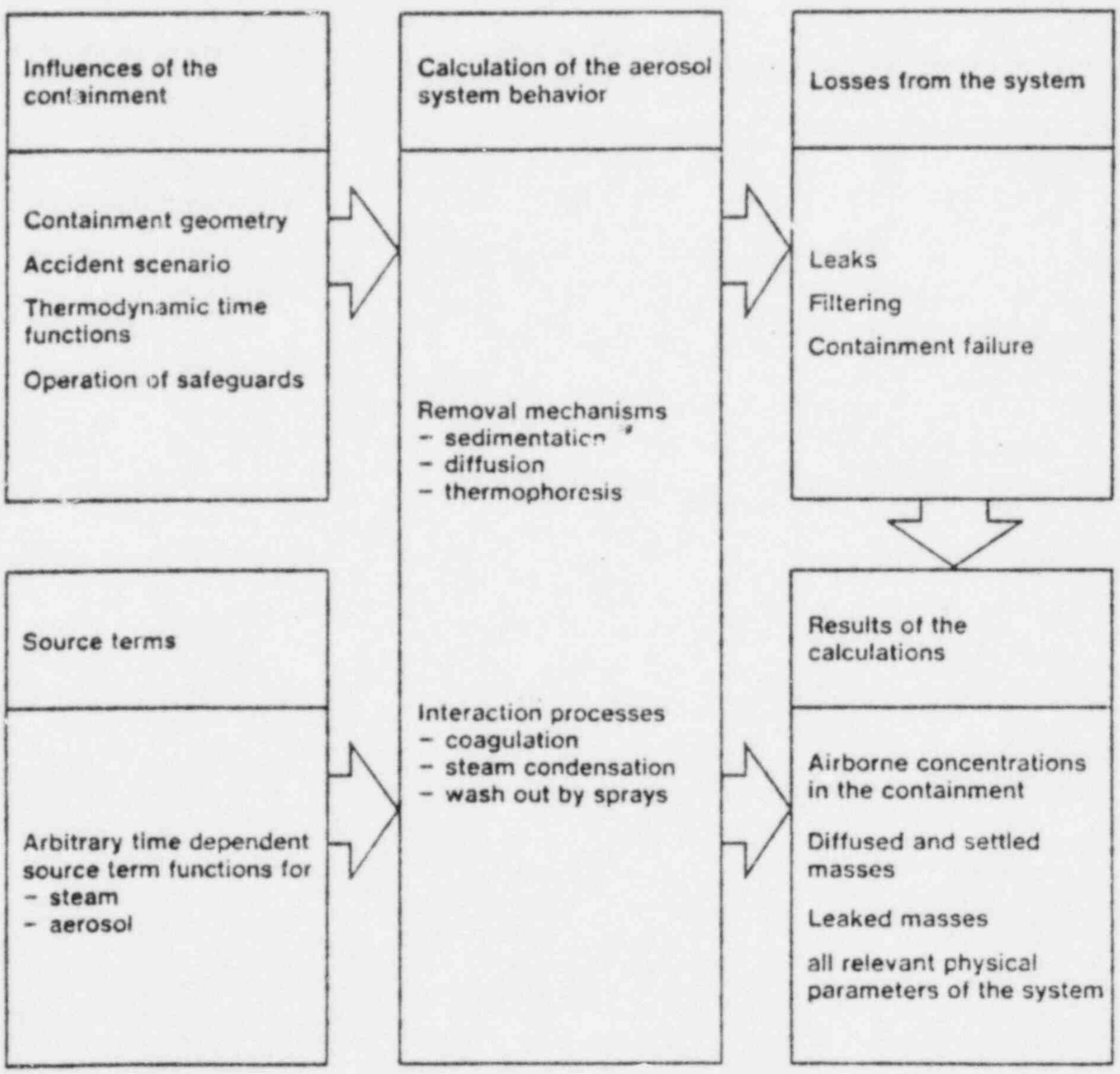
The NAUA Model and Computer Code

predict the removal of radioactive aerosols from the atmosphere of an LWR-containment during hypothetical core meltdown accidents.The model is based on microscale physical aerosol processes in a condensing atmosphere.The relevant parameters are determined experimentally.



Physical scheme of the NAUA computer code

Fig. 1

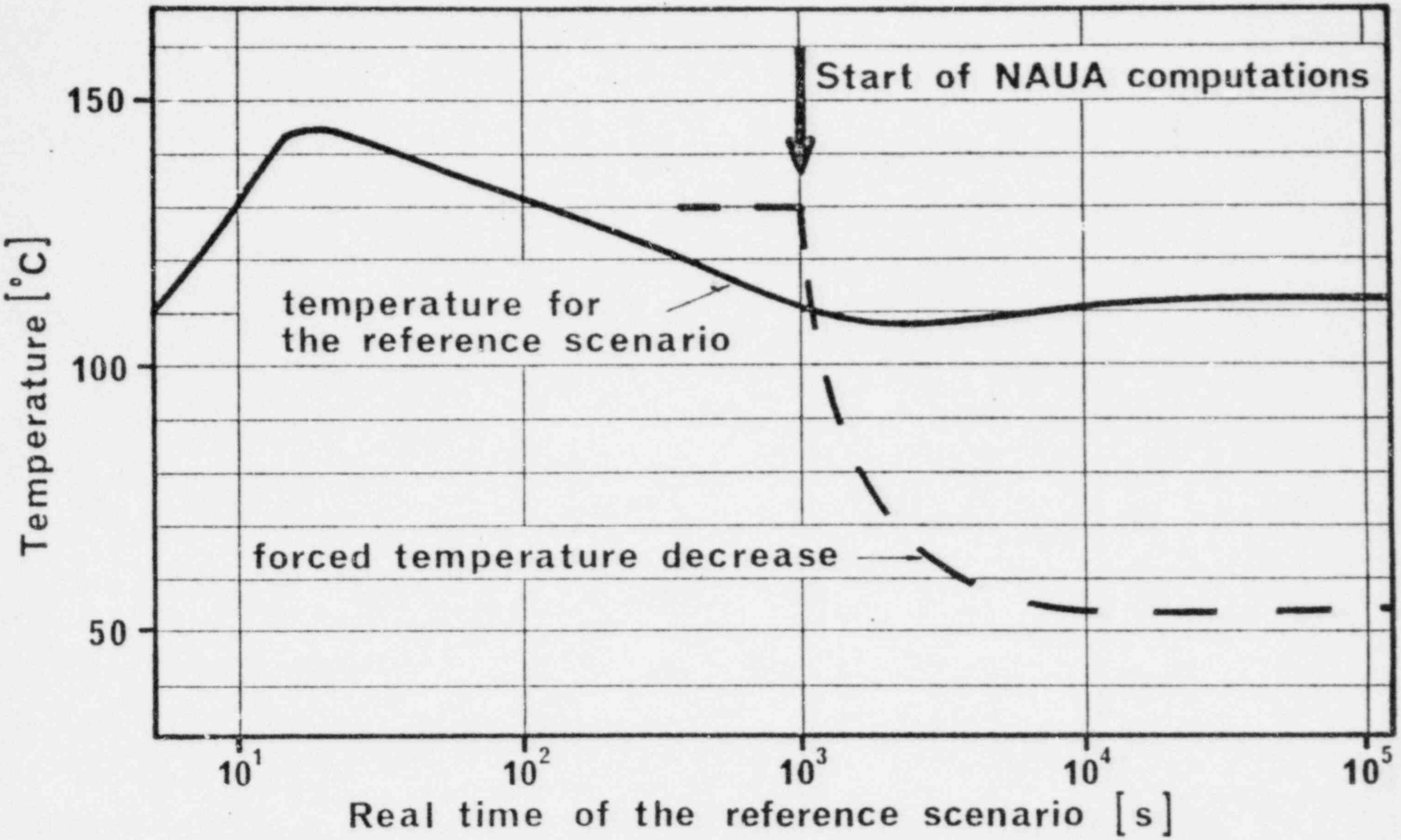


Fig. 2: Containment temperature function for the NAUA calculations

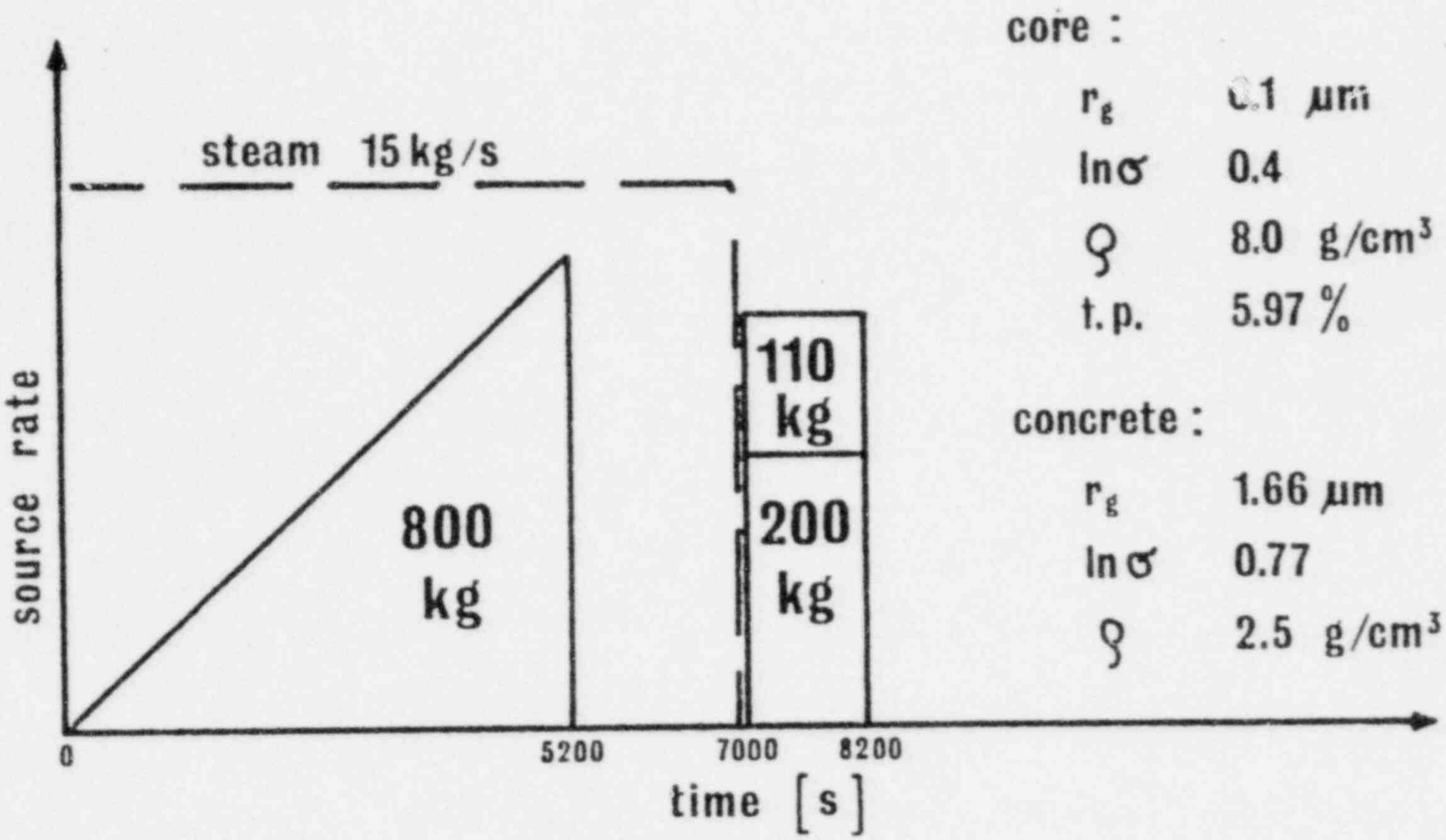


Fig. 3: Aerosol and steam sources for the NAUA calculations

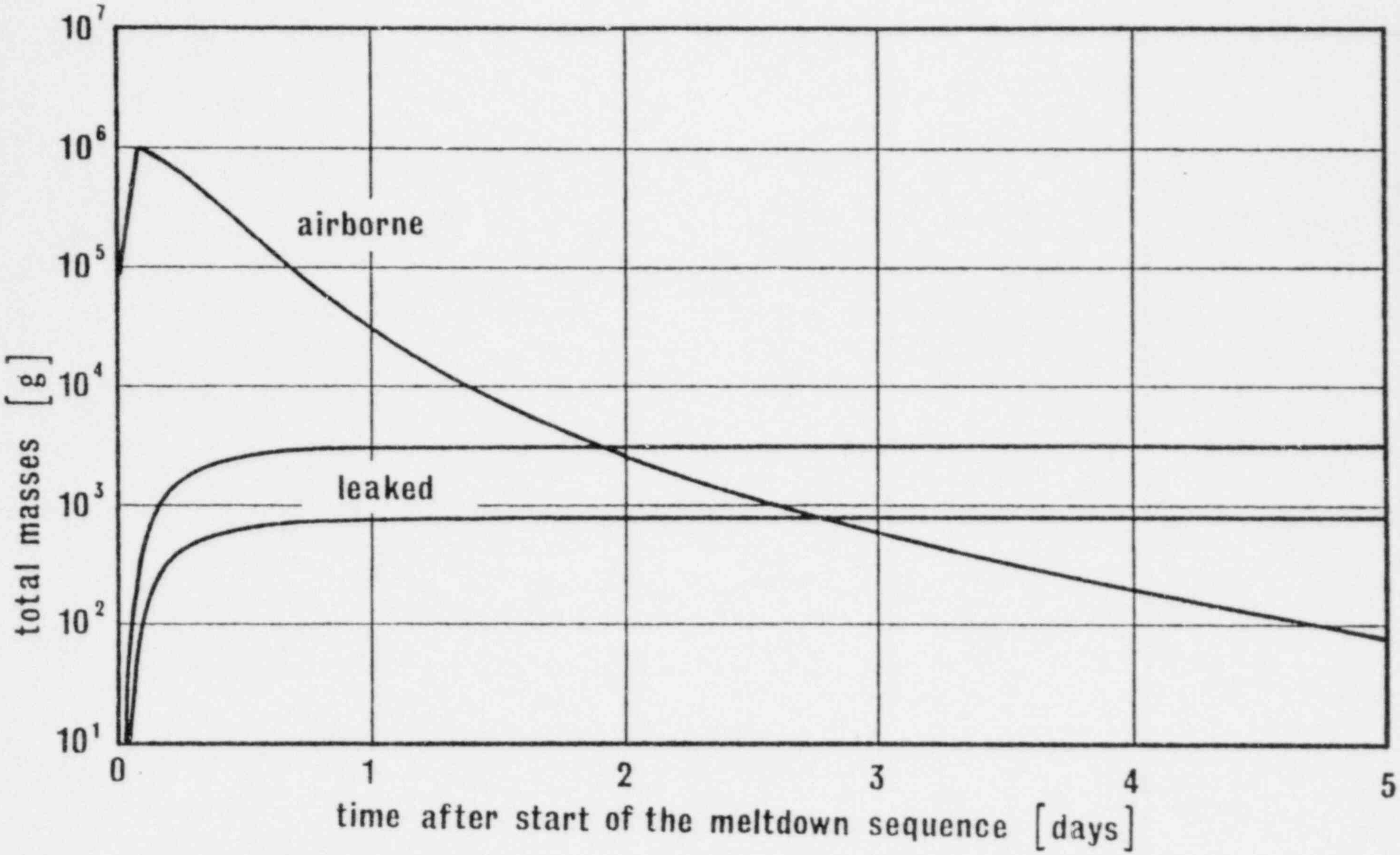


Fig. 4: Airborne mass in the containment and leaked mass.
Leak rates 0.25 and 1.0 % / day

total released fuel aerosol mass [kg]	leaked aerosol mass		
	without steam source [g]	with steam source [g]	with forced temperature decrease [g]
2000	1186	1146	—
1000	754	711	239
100	172	156	—

Table 1: Leaked aerosol mass for different scenarios