

Aerosol Release from a Hot Sodium Pool and Behaviour in Inert Gas Atmosphere

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ABSTRACT

In the KfK-NALA program, experiments were carried out on the subject of aerosol release from a contaminated sodium pool into inert gas atmosphere under various conditions. Besides the determination of retention factors for fuel and fission products, the sodium aerosol system was investigated and characterized, concerning aerosol generation (evaporation rate), particle size, mass concentration, and deposition behaviour. Pool temperatures were varied between 700 and 1000 K at different geometrical and convective conditions. Technical scale experiments with a 531-cm<sup>2</sup> pool surface area were performed at natural convection in a 2.2-m<sup>3</sup> heated vessel, as well as additional small scale experiments at forced convection and 38.5-cm<sup>2</sup> pool surface area.

A best-fit formula is given for the specific evaporation rate into a 400 K argon atmosphere. Approximative, the very convenient relation  $\dot{m}$  (kg/m<sup>2</sup>/h) = 0.1 p (mm Hg) was found. The sodium aerosol diameter lay between 0.6  $\mu$ m, less than 1 sec after production, and 2.5  $\mu$ m at maximum concentration. The deposition behaviour was characterized by very small quantities (< 1%) on the top cover and large quantities (> 80%) on the bottom cover of the vessel.

In the modeltheoretic studies with the PARADISEKO code, calculations were performed of the mass concentration, particle diameter and deposition behaviour. Agreement with the experimental values could not be achieved until a modulus was introduced to allow for turbulent deposition.

## 1. Introduction

In the Karlsruhe Nuclear Research Center-NALA program, experiments are carried out on the subject of aerosol release from a contaminated sodium pool into inert gas atmosphere under various conditions. It is mainly related to the release of fuel and fission products, but the determination of sodium evaporation rates and the characterization of the sodium aerosol system is an important by-product. In this paper, we focus on the results concerning sodium aerosols from technical scale experiments (NALA II, natural convection) and their interpretation. For completeness, results from laboratory scale experiments (NALA I, forced convection) are also given.

The program is related to the SNR 300 core catcher problem /1/ (pool temperatures from 700 to 1000 K, inert gas temperature 400 K). The applications of the results, however, are manifold. For more detailed information, the reader is referred to refs /2, 3/.

## 2. Description of the NALA II facility

A schematic drawing of the NALA II facility is shown in Fig. 1. The main components are a 2.2-m<sup>3</sup> heated vessel, a sodium pot of 26-cm i.d. with external heating devices (acetylene burner) to be flanged to the vessel bottom, inert gas supply, survey instrumentation, and aerosol measurement instrumentation. The vessel is usually filled with argon gas at normal pressure (1 bar) and heated up to 400 K; this temperature is kept constant during the whole experiment. In the sodium pot, sodium amounts of the order of 1 kg are heated up to the desired pool temperature in the range between 700 and 1000 K. By flame regulation, a temperature plateau is approximately achieved. The duration of external heating (0.7 to 3 h) is adjusted to the evaporation rate in such a way that ~10 % of the sodium pool evaporates. Temperatures were measured at six different locations in the vessel and at two locations in the sodium pot. During the period of external heating, all containment gas temperatures were practically the same. A typical diagram of pool and gas temperature versus time is shown in Fig. 2.

### 3. Sodium evaporation rate

Strong aerosol production is observed in the vicinity of the pool. Due to aerosol settling and vapor condensation, the evaporated sodium is deposited at the vessel walls, mainly as very fine droplets. Since we have a completely closed system (including aerosol sampling and aerosol trapping in the pressure regulating system), the total amount of evaporated sodium can be determined at the end of an experiment. Thus, each experiment provides integral information on the sodium evaporation rate.

The aim of the analysis is to determine a number  $\dot{m}$  for the average specific sodium evaporation rate for each of our experiments, to give a best-fit formula to describe the  $\dot{m}$  versus  $T$  relation for all of our numbers, and to give, if possible, a simple and convenient relation (i.e., by a scale factor) between  $\dot{m}$  and the vapor pressure  $p$ .

The averaging procedure which is necessary since we have no constant pool temperature is indicated in Fig. 2 and described in detail in ref. /2/. Average pool temperatures and average specific evaporation rates from seven NALA II experiments are shown in Fig. 3 and related to the vapour pressure.

The evaporation rate of a metal can be described by a correlation of the type

$$\log \dot{m} = A - \frac{B}{T} - 0.5 \log T$$

Our best-fit values are  $A = 8.062$  and  $B = 5426$ , valid for  $\dot{m}$  in units of  $\text{kg m}^{-2} \text{h}^{-1}$  and  $T$  in units of Kelvin. As illustrated in Fig. 3, the very convenient relation

$$\dot{m} (\text{kg m}^{-2} \text{h}^{-1}) \approx 0.1 \cdot p (\text{Torr})$$

can be used as a reasonable estimate of sodium evaporation rates at our conditions.

#### 4. Particle-size measurements on sodium aerosols

With an eight-stage Andersen impactor, particle-size measurements were performed on the sodium aerosols. The principal aim was to determine the time dependence of the median particle diameter (i.e., the 50 % mass diameter) over the whole run time. Fig. 4 presents a typical example. The plot shows the cumulative measured frequencies of the sodium aerosol mass distribution, in log-normal probability coordinates, at the start of a run ( $t = 15$  min) and at a late phase ( $t = 5$  hr). The mass distributions are characterized by the median mass diameter  $d_m$  ( $m = 50$  %) and the geometric standard deviation  $\sigma_g = d(84\%)/d(16\%)^{1/2}$ . For the example shown, we found:

$$t = 15 \text{ min: } d_m = 0.82 \text{ } \mu\text{m}; \sigma_g = 1.90.$$

$$t = 5 \text{ hr: } d_m = 2.03 \text{ } \mu\text{m}; \sigma_g = 2.20.$$

An earlier impactor measurement immediately after aerosol formation (i.e., within 1 sec.), yielded  $d_m = 0.61 \text{ } \mu\text{m}$  (see chapter 8)

In general, it was found that  $d_m$  increases rapidly from initial values less than  $1 \text{ } \mu\text{m}$  to maximum values of about  $2.5 \text{ } \mu\text{m}$ , then (after the source is turned off) declines very slowly; it is largely independent of the experimental conditions. Deposition processes become so effective that further increases in the 50 % value are prevented even at high mass concentrations (up to  $20 \text{ g Na/m}^3$ ).

#### 5. Aerosol mass concentration

The condensation of vaporized sodium and thus the formation of an aerosol take place in the immediate vicinity of the pool surface. At temperatures of about  $300 \text{ } ^\circ\text{C}$  and higher, mist formation can be clearly seen above the surface. As the temperature rises, the aerosol formation rate increases strongly. The concentration reaches an equilibrium (maximum) value when formation is balanced by separation processes. After the source is turned off, the concentration initially falls relatively fast, but the decline later slows down markedly.

The sodium aerosol mass concentration was measured over a span of up to 10

hours. Fig. 5 shows the time dependence of the mass concentration for the example of trial T 7. A significant feature of the decline in concentration is that it can be represented approximately by two exponential functions. Maximum concentrations up to 20 g Na/m<sup>3</sup> were measured.

## 6. Deposits on walls

The deposition of sodium aerosols on the walls follows a recurrent pattern which can be seen visually and investigated quantitatively after the tank is opened and top cover, cylindrical section and bottom cover are dismantled. Very little sodium is deposited on the top cover (about 0.5 % of the quantity released), whether the cover is heated or not. Visible traces occur mainly on edges and projecting corners. Smooth vertical wall surfaces have only slightly more deposits than the top cover. Most of the "wall deposits" in the cylindrical section come from small horizontal surfaces. The total amount is between 12 % and 18 %. The bottom cover holds the bulk of the deposits, always over 80 %.

Collecting plates which were mounted horizontally barely below the surface of the top cover bore deposits similar to those on the corresponding sides of collecting plates near the bottom cover. In contrast, the back sides had almost no deposits.

## 7. Numerical simulation of aerosol behaviour

The NALA II experiments are especially well suited to studies with aerosol behaviour codes. The boundary conditions (geometry, temperature, pressure, gas properties) and the experimental results used for comparison (aerosol mass concentration, particle size, quantities deposited) are known relatively exactly. The sodium aerosols can be viewed as spherical. The partial vapor pressure of sodium in the tank volume at 130 °C is so low that all the sodium vaporized can be considered as converted to aerosols in the immediate vicinity of the pool. Calculations were done with the PARDISEKO code, initially with

version III b /4/. These calculations in which the aerosol processes of coagulation, diffusion and sedimentation are simulated give a maximum mass concentration that is a factor of about 3 too large and a concentration decline that is too slow. The typical "knee" in the measured time dependence is not reproduced. In the calculations where "thermophoresis" is used as a parameter in fitting to the mass-concentration curve, the deposition on the wall is often overestimated and the particle diameter found is too large.

Finally, studies with turbulent deposition of particles on the top cover, wall, and bottom cover surfaces, using the computational procedure given by Sehmel /5/, reproduced the time dependence of the aerosol mass concentration, but also simulated the mean particle size. These studies ultimately resulted in the version IV of the PARDISEKO code /6/. In Fig. 5, an example of a mass concentration curve (experiment versus theory) is shown.

#### 8. Additional small scale experiments

Different types of small scale experiments (NALA I) were performed, mainly for the purpose of variation of pool temperature, gas convection, and geometry. Two characteristic setups are described in the following:

I. In a small, cylindrical (7-cm-diam) stainless steel vessel, sodium ( $\approx 100$  g) was heated up to the desired temperature in an oven. The released amounts were transported by an inert gas stream (forced convection) at isothermal conditions into cold traps. Temperatures between 700 and 1160 K (sodium boiling) were achieved.

II. An open vessel, similar to the A type, was installed in a glove box with nitrogen atmosphere. By electrical heating, sodium temperatures up to 840 K were achieved. The evaporated sodium was released into the ambient nitrogen atmosphere ( $T \approx 325$  K) and sucked from there via a funnel into cold traps and filters for complete deposition. The purpose of this series was to study the release rates at seminatural convection in the vicinity of the pool and at large temperature gradients in preparation of the NALA II experiments.

A sketch of both setups is given in Fig. 6. Among others, evaporation rates were studied at various convective conditions (see refs. 2, 3, 7), and particle sizes were measured immediately after generation (see chapter 4). For boiling sodium, evaporation rates of the order of 85 kg/m<sup>2</sup>/h were found.

#### References

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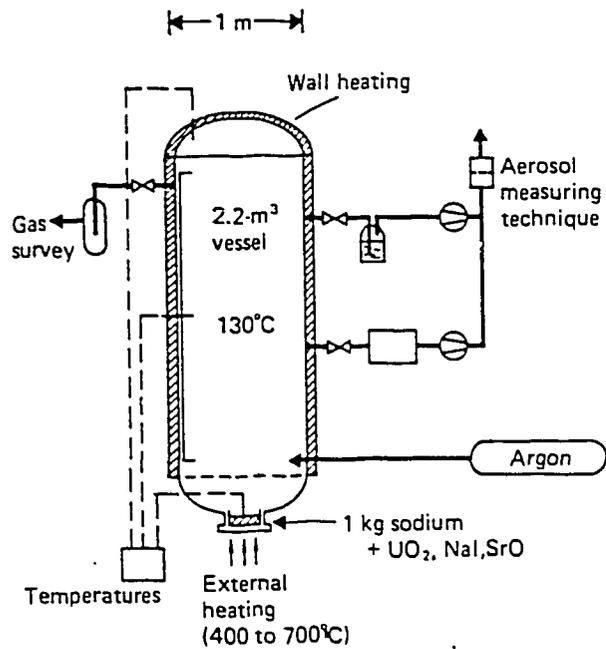


Fig. 1 Schematic diagram of the NALA II facility

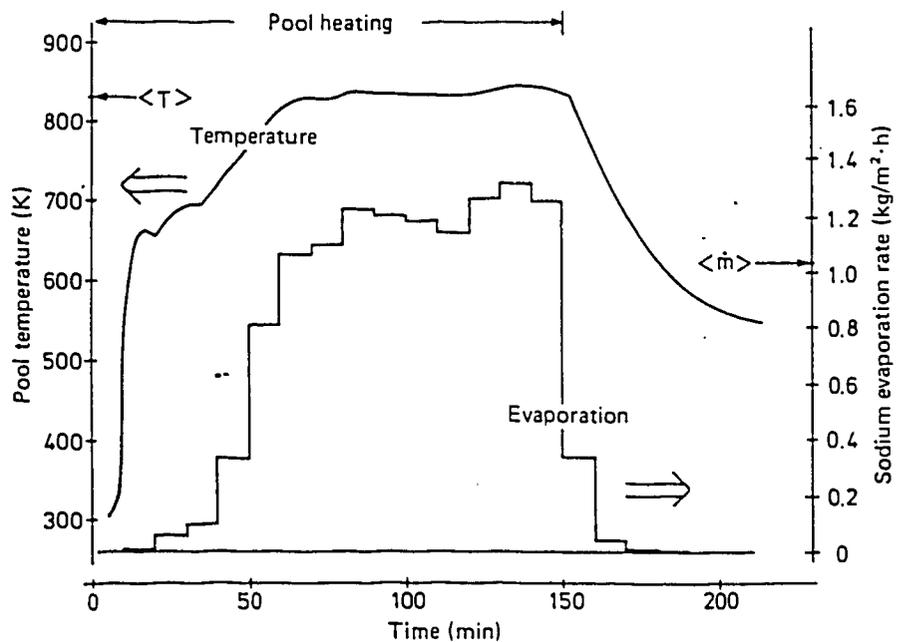


Fig. 2 Sodium pool temperature (measured) and sodium evaporation rate (stepwise calculation) for experiment NALA - T4. Indicated are the time of external pool heating, the average pool temperature  $\langle T \rangle$  and the average sodium evaporation rate  $\langle \dot{m} \rangle$ .

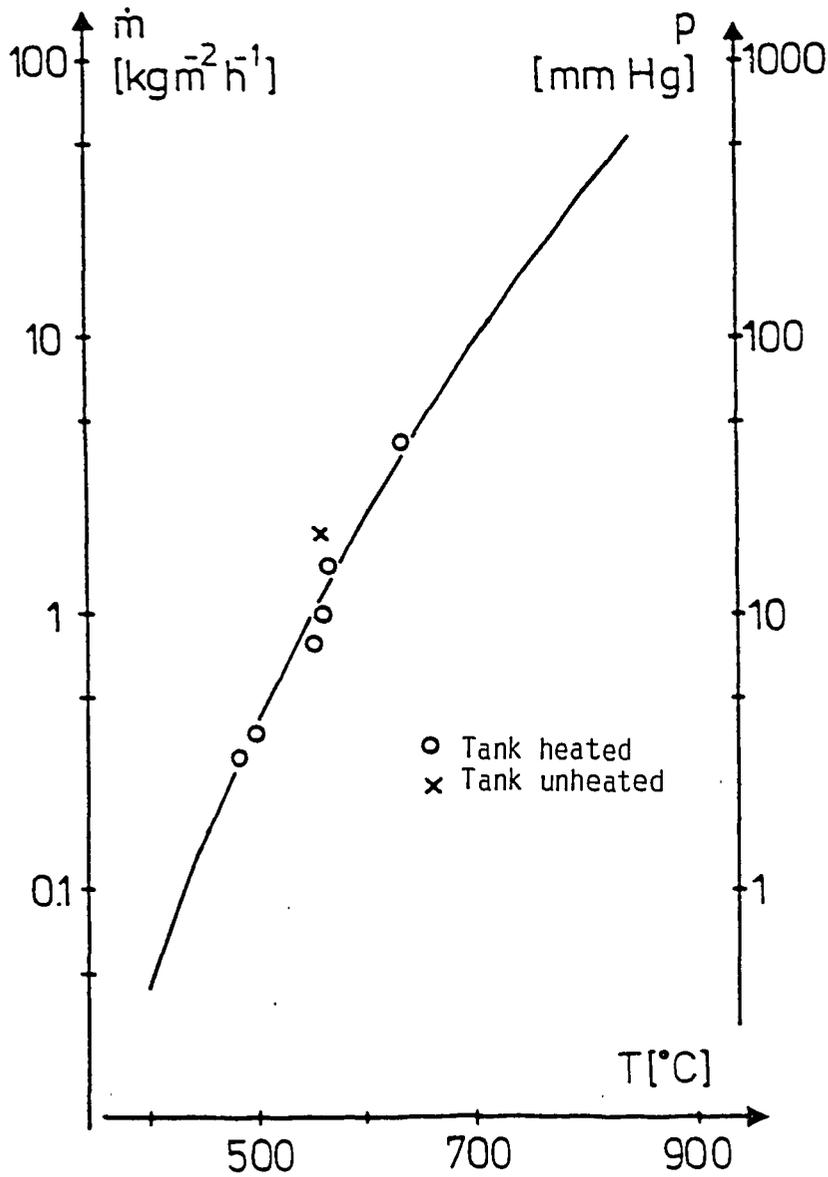


Fig. 3 Sodium evaporation rate

The measured points (from seven experiments) are to be read against the left-hand scale. The solid curve shows the sodium vapour pressure and is to be read against the left-hand scale. The scales are displaced just one order of magnitude against each other.

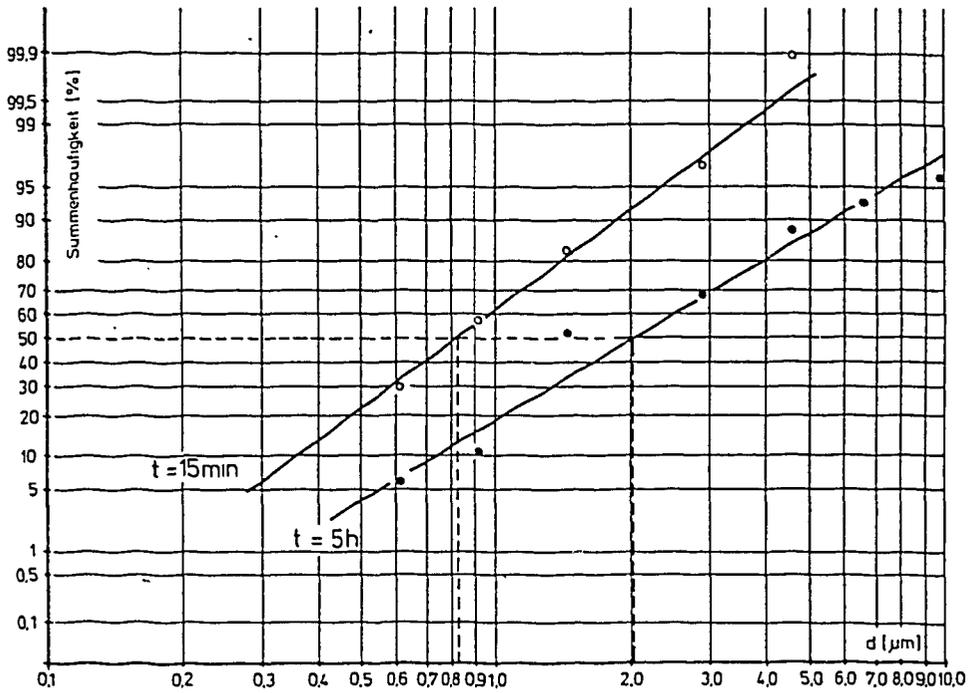


Fig. 4 Sodium aerosol size distribution, measured with an eight-stage Andersen impactor during experiment NALA - T5.  
 $t$  = time after start of external pool heating

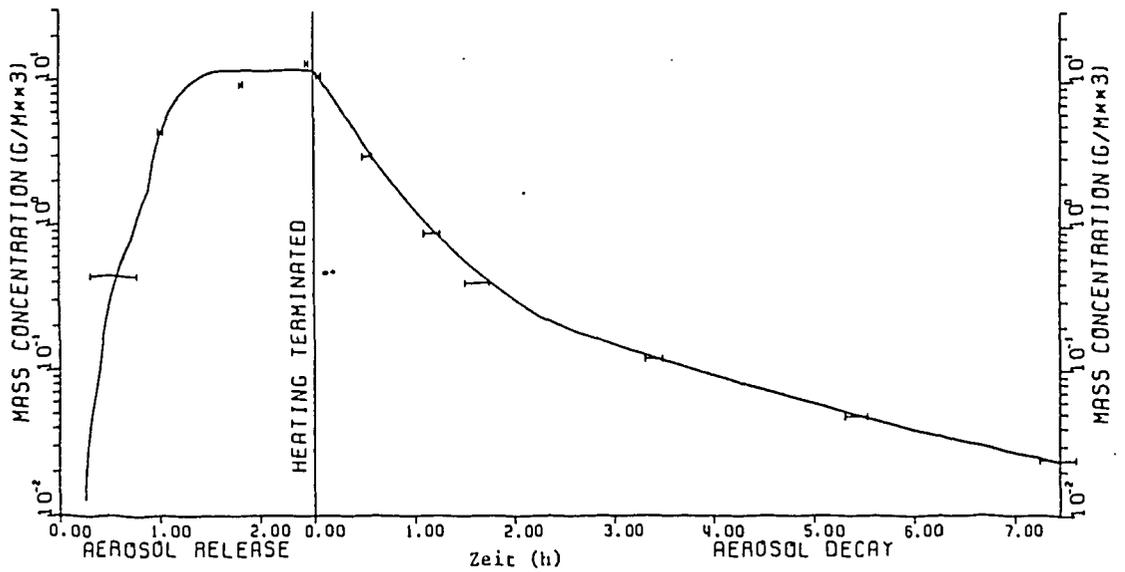
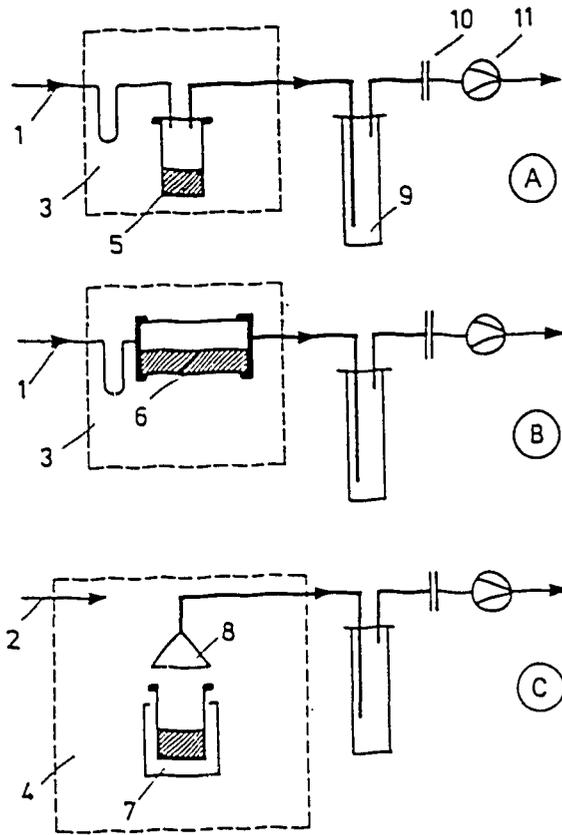


Fig. 5

Sodium aerosol mass concentration: Experiment versus PARDISEKO IV calculation



1. Argon carrier gas, preheated
2. Nitrogen supply
3. Oven ( $T_{\max} = 1400 \text{ K}$ )
4. Glove box
5. Stainless steel vessel, 7 cm in diameter, 10 cm high, with 100 g sodium
6. Stainless steel cylinder, 6 cm in diameter, 20 cm long, with 150 g sodium
7. Open stainless steel vessel with electrical heating, same dimensions as item 5
8. Funnel
9. Cold trap
10. Filter (0.2- $\mu\text{m}$  pore size)
11. Pump and gas metering system

Fig. 6 Schematic diagram of small scale experiments: Type A, B under forced convection at isothermal conditions, type C under quasi-natural convection in a glovebox set-up.