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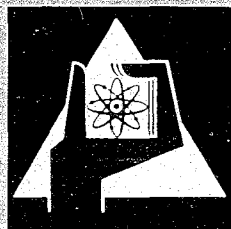
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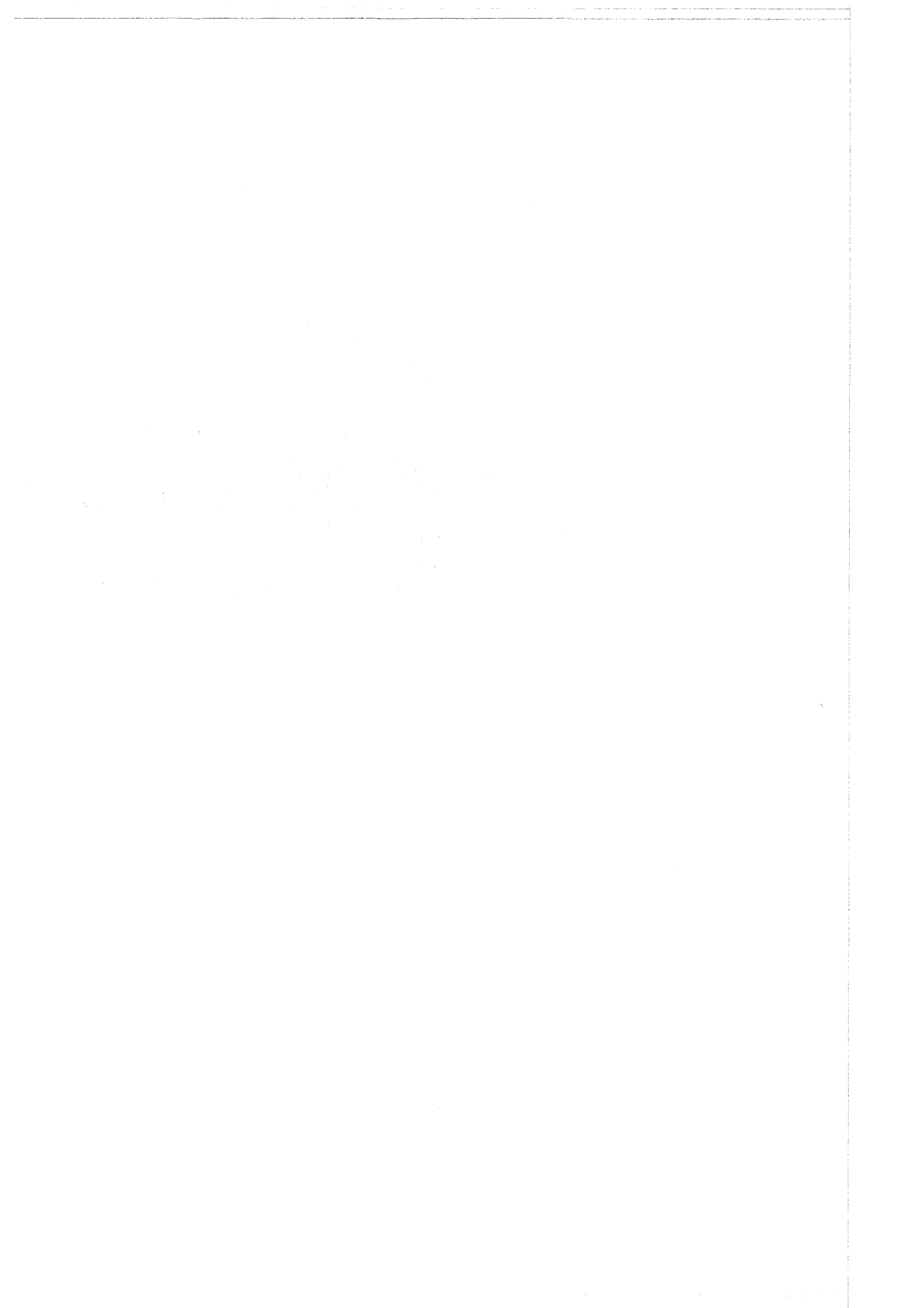
The Karlsruhe Research Program on Nuclear Aerosols and its
Relation to the Plutonium Hazard of Fast Sodium Reactors

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1. INTRODUCTION

Reactor siting is mainly a problem of activity release due to a major accident. In estimating the accidental activity release and assessing the hazard to the public, accident and activity release models are used. Many parameters which are not well known or even not well understood enter the calculations. Since data are lacking numerous conservative assumptions are made. In particular, the physical and chemical nature of the activity carrying material released from the reactor fuel, its behaviour in an after accident containment atmosphere, its leakage and transport characteristics, and the potential radiation exposure still bear many unresolved questions. The importance of the subject is illustrated by the number and size of research programs conducted in various countries on the various aspects of activity release.

The Karlsruhe Research Program on Nuclear Aerosols in the present phase is directed at some specific problems connected with the accidental activity release of fast breeder reactors. In particular, in sodium cooled fast breeders some specific questions exist in respect to activity release and to activity release reducing engineered safeguards. Better understanding of the phenomena involved would certainly give also better insight in the unresolved questions of accidental activity release from thermal reactors.

This paper discusses

- characteristics of activity release models
- the role of aerosol formation in activity release
- the objectives of the Karlsruhe aerosol program

2. CHARACTERISTICS OF ACTIVITY RELEASE MODELS

The inventory of radioactive material in a reactor core could be up to 10^8 Ci depending on isotope and burn up. The maximum permissible free handling activity could be down to 10^{-6} Ci depending on isotope and burn up. Therefore, 14 decades in activity is the range which must be

handled in assessing activity release in core destructive accidents.

It depends on various parameters as reactor design, the design of the containment system and meteorological conditions how much of the activity released from the fuel in an accident could reach a person being in the vicinity of the reactor. Most important problems, however, are the

- function of activity release from the core and
- decontamination of the containment atmosphere from airborne activity

At present most reactor accident analysts use what may be called the "evaporization model" in describing the amount and time function of activity release from reactor fuel. This model in his most simple form assumes that fission products and fuel material can escape from the core into the containment atmosphere according to the fuel temperature encountered in the accident and the corresponding vapor pressure of the fission product isotope under consideration.

Therefore, in core destructive accidents like core meltdown due to loss of coolant of a thermal reactor the part of activity inventory which is assumed to be released from the fuel can be calculated in terms of the percentage of core melting since the boiling point of most fission products, particularly the important ones in respect to radiation burden, do not exceed the melting point of UO_2 . More or less all important fission products are released from the fuel after reaching the melting point. In accidents like core vaporization due to a reactivity excursion of a fast reactor the activity release is even easier to predict. Since essentially all elements or compounds existing in reactor fuel after burn up have boiling points lower than the UO_2 boiling point all fission products, fuel isotopes, cladding and coolant elements will be vaporized and released to the containment atmosphere.

It can be concluded that in the "evaporization model" the percentage of released radioactive material is fairly high and probably overestimated. This is certainly a conservative but also a pessimistic approach.

The "evaporization model" can describe the primary activity release "on the safe side". However, it lacks at an important point, namely, it does not take into account the physical form of the released material which is responsible for a number of decontaminating processes taking place in the atmosphere. Here a better approach, what we call the "aerosol model", can improve the situation. This model is based on the following arguments:

- Large accidents in nuclear reactors set free very large amounts of radioactive material which must stay airborne for long times (hours) to leak through the containment barriers and present a hazard to the environment
- the released material could be in the form of gases, vapors and aerosols. Since vapors behave very much like gases if their droplets are small enough, and since some vapors could be considered as aerosols, the released material exists essentially in the form of gases and aerosols
- the release of gases out of the reactor plant can be predicted fairly accurate. The problems to resolve lay in the understanding of the transport characteristics of nuclear aerosols generated in core destructive accidents and being suspended in the containment atmosphere.

The aerosol model provides several advantages which could lead to a more

realistic description of activity release. In the aerosol model we do not ask how much active material is released from the fuel, rather we ask how much active material can stay airborne in the containment atmosphere during the accident. Therefore, we do not ask for release fractions of specific fission products which will be subject of doubt as long as accident models are not sufficiently verified. We rather ask for aerosol behaviour and aerosol parameters which provide much better access to experimental investigation and justification. Furthermore, the aerosol model illuminates which parameters are of importance in the attempt to reduce the activity release in an accident. For instance, the containment volume is direct proportional to the airborne aerosol mass, and therefore, activity. Minimizing the free containment volume can help reducing the gross activity release considerably. Studying the after accident aerosols would lead also to optimized engineered safeguards as filter systems, spray systems and others.

3. ACTIVITY RELEASE VIA FORMATION OF AEROSOLS IN FAST REACTOR ACCIDENTS

Accidental activity release in fast reactors differs from that of thermal reactors because of two aspects. First, the initiation and course of large accidents in fast reactors are different, second, the amount of radioactive inventory capable to be released into the containment atmosphere is different. At present large accidents in fast reactors are assumed to produce reactivity excursions with subsequent melting and vaporization of reactor fuel and of other core materials. Therefore, activity release from the core is nearly complete due to vaporization of all core material and depends only on the percentage of core vaporization. Consequently also solid materials as high melting oxides can be released.

Since almost all core materials can be released from the core there is no sense to define any release fractions for any specific radioactive elements. Rather we have to ask how the released material behaves after the vaporization. Depending on pressure, temperature and other conditions it will certainly recondense and form aerosols which then can be subject of various processes as coagulation, diffusion, thermophoresis, sedimentation and plate-out. Fortunately, most of these processes tend to decrease the airborne aerosol concentration, i.e. the containment atmosphere has a self-cleaning effect. However, because these decontamination processes depend on many parameters which are not well known, the amount and the time function of these air cleaning processes can not be predicted reasonably.

A number of aerosol parameters must be known to estimate activity transport in containment systems after accidents. First of all the primary particle size distribution is important. It depends strongly on the energy inserted into the fuel during vaporization. Particles created by recondensation in most cases are observed to be spherical. Soon after production of the primary particles coagulation takes place forming larger agglomerats. These agglomerats differ in size, form, weight, porosity etc. depending on the primary particle size and on the conditions under which the agglomerats are formed. The particle coagulation depends strongly on the particle concentration. Therefore, most important parameters in the description of aerosol formation and behaviour after a large accident with core vaporization are

- primary particle size distribution and their time behaviour
- primary particle concentration and their time behaviour
- particle coagulation

Knowing these parameters most other processes as sedimentation or diffusion can be estimated. It is mentioned that the various accident conditions as presence of vapors and gases, gas temperature, pressure, temperature gradients, gas flow etc. influence the aerosol behaviour considerably. Because of the presence of various elements in the gas phase after vaporization, various chemically different aerosols can be formed which may coagulate to mixed aerosols. Since most of these effects cannot be predicted reliably experimental studies are necessary to investigate the importance of the various postulated effects and conditions.

The second aspect in which accidental activity release of fast reactors differs from that of thermal reactors is the radioactive inventory. Fast reactors can contain 8 times more Plutonium. This emphasizes the importance of the accidental release of solid fuel and fission products due to a core vaporization accident. Plutonium could be a major hazard to the public in case of such an accident, if all the Plutonium would be released. This is illustrated by figure 1 which shows the contribution to the environmental accident dose in absence of any activity decreasing engineered safeguard.¹ Plutonium as a bone seeker and taking its way through the lung makes the major contributions to these doses. It is mentioned, however, that plate-out factors standing for the selfcleaning effect of the containment atmosphere are of large influence and are assumed pessimistic. As has been shown earlier [1] Plutonium doses can be reduced up to a factor of 10^4 if the aerosol model with aerosol parameters which are reasonably extrapolated but not yet experimentally or theoretically confirmed and justified, is applied.

Accident dose calculations are rather complex because of numerous reactor and site data involved. They are significant only if they are related to a real reactor plant or reactor design. Therefore, the answer to the problem which isotope is in the case of accidental release the limiting factor in siting may vary with reactor design, accident analysis and site conditions. In our example the doses are calculated taking into account all fuel and fission product isotopes produced during operation of a 300 MWe sodium cooled fast reactor. All doses are calculated according to the ICRP recommendations [2]. Only in the case of the lung dose a shorter biological half time for PuO_2 reported in [3] was used.

Whether the bone or the lung is the critical organ for Plutonium aerosol inhalation depends on several aerosol parameters. First of all the element composition in the aerosol is important since not only just PuO_2 or Pu aerosols will be created. Much more probable is the formation of mixed aerosols containing several elements of the vaporized core material. The degree of mixing depends on coagulation which depends on primary particle

¹ Dose calculations are based on following assumptions:

reactor power: 750 MWth, load factor: 0,7, operation time: 430 d

	halogens	solids	volatile solids	noble gases
release fractions	10%	1%	50%	100%
plate-out half time	1h	10h	1h	

double containment (leak rate 1: 50%/d, leak rate 2: 0,5%/d), stack release

concentration. Mixed aerosols may be much more soluble in the body fluid than insoluble metal oxides. Furthermore particle size influences the internal body transport of inhaled particles. The probability of entering the blood stream will increase with decreasing particle size.

Another aspect is the off-gas filter system. We do not know very much how filter systems behave if hundreds of kilograms of dust, i.a. the vaporized core material, becomes airborne, and clogs and heats up (because of decay heat) the filter system. The high efficiency filter may encounter a considerable loss of efficiency. Again particle size and other aerosol parameter are important. In view of these considerations we did not take into account filter factors in the dose calculations of Fig. 1 although a reasonable factor between 90 to 99 percent may be justified even for long term availability and efficiency of off-gas filters. These factors could drop the bone and lung dose down to the external dose which is mainly due to irradiation by noble gases.

4. THE KARLSRUHE AEROSOL RESEARCH PROGRAM

4. 1. Status of the problem

Nuclear aerosols, i.e. aerosols generated in large core destructive reactor accidents have not been studied very much in the past. This is due to a number of reasons, to mention a few

- Iodine is the most important fission product to be released in a thermal reactor accident. Most Iodine escapes from the failed fuel in vapor form creating aerosols only if certain conditions of the containment atmosphere are given
- large accidents in thermal reactor cores produce relatively little amounts of nuclear aerosols
- since the activity is the important factor most release studies concentrate on the measurement of activity transport characteristics rather than on the characteristics of the transported material as state, particle form, particle size and others.

Yet the accident analysis of large fast reactors and the growing use of Plutonium has put emphasis on the importance of nuclear aerosols. Here, as has been explained in the foregoing chapters, Plutonium bearing aerosols are of main interest.

Several investigators have attempted to determine the aerosol characteristics of airborne Plutonium. CARTER [4], STEWART [5] and ETTINGER [6] studied the aerosol produced from burning Plutonium. They found mass median diameters around 1 μ to 16 μ [4] [5] and 0,03 - 0,13 μ [6]. The aerosols were almost spherical. No chains were observed. SHERWOOD [7], MOSS [8] and BRUNSKILL [9] studied Plutonium aerosols from radiochemical laboratories. Here mass median diameters of 0,14 to 0,65 μ [8] and aerodynamic diameter of 5 to 20 μ [9] were found. No information on aerosol concentration, particle density or mass concentration, particle density or mass concentration was determined in these measurements which could be a tool for the estimation of Plutonium aerosol behaviour as function of large core destructive accidents.

Besides the large discrepancy in measured particle size little information can be drawn from these measurements for the accidental release problem because of the difference in the aerosol production process. It is known since a long time that aerosol properties, particularly particle size and form, depend on the energy involved in the production process. Other parameters as temperature, pressure etc. can play a role too. Therefore, the method of aerosol production is of high importance in this context. Experiments carried out by KARIORIS and co-workers [10] confirmed the influence of energy on particle size. They found decrease in particle size of metal oxide aerosols produced by exploding metal wires with increasing voltage of the capacitor bank. Unfortunately no attempt was made to evaluate the electrical energy actually introduced into the metal wire which, of course, is only a small percentage of the electrical energy available in the capacitor bank.

There is information on the particle size of metal oxides also coming from industrial process as ore mining, welding etc. Here, again a tendency to small particles of the submicron range $< 0,5 \mu$ can be observed [11]. However, a direct information on the particle size of Plutonium bearing aerosols can not be deduced from all these experiments, because of unknown parameters as

- aerosol producing energy
- aerosol mass density
- particle concentration
- mass concentration

Principally, metallic dusts and fumes can have particle sizes between molecular diameters around $0,001 \mu$ up to 100μ . It is therefore quite unknown at present what sizes Plutonium aerosols created in nuclear accidents could have although small particles below 1μ are more probable than larger ones.

Unfortunately data on the other important aerosol parameters as particle density and particle concentration are not available. From the theory of coagulation we know that high concentrations of particles are not stable. Concentrations higher than 10^7 particles/cm³ may stay airborne only a few minutes. Although we probably will have lower concentrations, it still has to be studied what the concentrations could be because of the irregular forms (chains, agglomerats etc.) the aerosols actually will have. The influence of irregular particle forms on mass concentration under typical accident conditions has to be studied.

On deposition, diffusion, thermophoresis and other decontamination processes well established theories are available which, however, in most cases are valid for spherical particles only. Therefore, for irregular particles experimental investigations are necessary to provide sufficient data on the effectiveness of these processes in decontaminating a containment atmosphere.

4. 2. Objectives of the program

In view of the foregoing considerations the Karlsruhe aerosol program is aimed to provide information on the following subjects

- particle size of fuel and fission product aerosols as function of energy involved and as function of other important accident conditions. Upper and lower limits of particle sizes shall be established to give accident analysts a guide if the accident conditions assumed are changed,
- particle and mass concentration as function of vaporized material, energy and accident conditions. Upper and lower limits shall be established to provide information for the assessment of the maximum transported aerosol mass in a reactor containment system. Direct or indirect measurement of particle density shall be made,
- particle chemical composition as function of the accident condition. Because of the expected mixed aerosols the amount of the chemical elements involved and the corresponding specific activity will be determined,
- time function of the aerosol mass concentrations to estimate the decontamination processes taking place after the accident in the containment atmosphere. Variation of typical accident parameter as temperature, gas, gas volume, will help understand the mechanism involved,
- aerosol transport and decontamination studies from the theoretical point of view. Modelling of most important processes shall make the phenomena computable.

The overall objective of the Karlsruhe research program on nuclear aerosols is the complete understanding and the reliable description of activity release and transport in large nuclear reactor accidents. Since nuclear aerosols play an important role in the activity release the understanding of aerosol behaviour will contribute considerably to the understanding of accidental activity release.

4. 3. Program outline

The first phase of the program is concerned with fuel aerosols and their production and measurement at accidentlike conditions. In a preoperational research and development program the detailed outline of the experimental program and of the necessary facilities were established and supporting theoretical studies were carried out. Since the aerosol formation process is of important influence on particle size and other aerosol parameter three aerosol production techniques are employed

- exploding wire technique
- resistance heating
- burning

The first aerosol production method used is the exploding wire technique. A small pin of UO_2 is preheated to produce electrical conductivity. Then electrical energy is inserted by discharge of a bank of capacitors. The electrical energy available is 54 kWsec with time constants of discharge around 1 msec. Both parameters are variable. This allows for the investigation of energy-time effects according to data evolving from the reactor accident analysis.

The second aerosol production method is based on the well known electrical heating of solid material in a boat. To avoid any influence of the boat material different materials as Tungsten, Tantalcarbide will be compared. Both methods will be used in the same aerosol production chamber on top of the main vessel and comparison of both methods will show how much the aerosol production process influences particle size and form.

Aerosol measurement takes place in a $2m^3$ vessel (main vessel) which is equipped with various devices based on the following methods

- sedimentation sampling
- thermal precipitation
- electrostatic precipitation
- optical counting
- filter column
- filter pack
- scintillation spectral analyzer

The hole particle size range (from 100μ to $0,001 \mu$) is covered by these instruments. The thermal precipitator collects particles of all sizes from 10μ down to $0,001 \mu$, but needs long times in case of small concentrations to collect statistically sufficient particle populations. The electrical precipitator also collects essentially all particles but allows for better differential sampling. Samples are photographed by light and electron microscope and sized on the basis of equivalent area (projected diameter). The large particle sizes and the particle concentrations are measured by optical counters (Bausch & Lomb). An improved version of present optical counters using a laser instead of light beam is currently under development at Battelle, Frankfurt. It will be available to the program in the near future. Filter methods are used as integral methods for mass concentration measurement and, hopefully, for differential mass concentration measurement. The hole range of expected particle concentrations from 10^6 particles/cm³ down to 10 particles/cm³ can be measured. The scintillation spectral analyzer which was first described by BINEK and co-workers [12] and which is under further development and improvement at Sartorius, Göttingen will be used to differentiate the various chemical elements in mixed aerosols and to measure their particle size.

There are four test series of the experimental program in the first phase:

Test series No. 1 consists of test runs of the electrical and preheating equipment, calibration of energy and insertion time, and tests of the various aerosol measurement techniques.

Test series No. 2 consists of the production of UO_2 aerosols by small excess energies² in air and measurement of particle size and concentration as function of time.

Test series No. 3 consists of the production of UO_2 aerosols by various excess energies² in N_2 , Ar and He and vapors including sodium vapor with normal and elevated temperatures up to $200^\circ C$. Measurement of particle size and concentration as function of time will be performed and the creation of mixed aerosols will be investigated.

Test series No. 4 consists of production of UO_2 aerosols by other methods as resistance heating or induction heating. Comparison will be made to the aerosol produced in test series No. 3 to evaluate influence of aerosol production on aerosol properties.

² excess energy is defined as the energy which is introduced into the probe to exceed the melting point. This energy is responsible for release of particles from the probe

The main objective of the first phase of the program is to demonstrate the validity of the aerosol model described and to verify the upper limits of mass concentrations of fuel aerosols being stable in a typical fast reactor accident containment atmosphere and their time behaviour.

The second phase of the program is concerned with other aerosols than those consisting of fuel material. In particular those fission product aerosols will be investigated which are most hazardous to the public. Also nuclear aerosols produced in large accidents of thermal reactors will be studied. Aerosols produced by melt-down of fuel in shipping casks and aerosols produced by burning will also be studied.

As an integrated effort computer codes are developed [13] and will be improved by tracing the most important problems to be resolved.

4. 4. Description of facilities

The facility built for the experiments in phase 1 is called "Teststand für die Untersuchung Nuklearer Aerosole (TUNA)" and shown in Fig. 2. The 2m³ vessel consists of stainless steel for better cleaning possibility. The aerosol production chamber with the preheating device for UO₂ pins (Fig. 3) sits on top of the vessel. Low inductance cables connect the capacitor bank. Several flanges allow the connection of the aerosol measurement equipment and of the detectors for temperature, humidity, pressure, temperature gradient measurement. Very low leakage even at higher pressures can be achieved. The filling with gases and vapors including sodium vapor and steam is possible. Heating of the vessel is possible up to 200° C. The capacitor bank has an electrical energy of 54 kWsec. This energy is sufficient to vaporize about 1 cm³ of UO₂. Discharge with several time constants between 100 μsec and 10 msec is possible depending on the resistance characteristics of the probe to be vaporized and other electrical properties of the facility. Penetrations are designed for high voltage. Aerosol production by electrical resistance heating can be done in the aerosol chamber as well.

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SUMMARY

The activity release from nuclear reactor cores due to large accidents is discussed and the role of aerosol formation and aerosol behaviour after large accidents is described. For the particular case of fast reactors the release of Plutonium as aerosol is analyzed and the importance and possibly limiting nature for reactor siting is shown.

Experimental program and test facilities of the Karlsruhe research program on nuclear aerosols are described and the objectives of the program discusses.

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FIG. 1

LARGE ACCIDENT DOSE TO DIFFERENT ORGANS
OF TYPICAL FAST SODIUM REACTOR

FIG. 2

TUNA

FIG. 3

PREHEATING DEVICE FOR UO_2 PINS

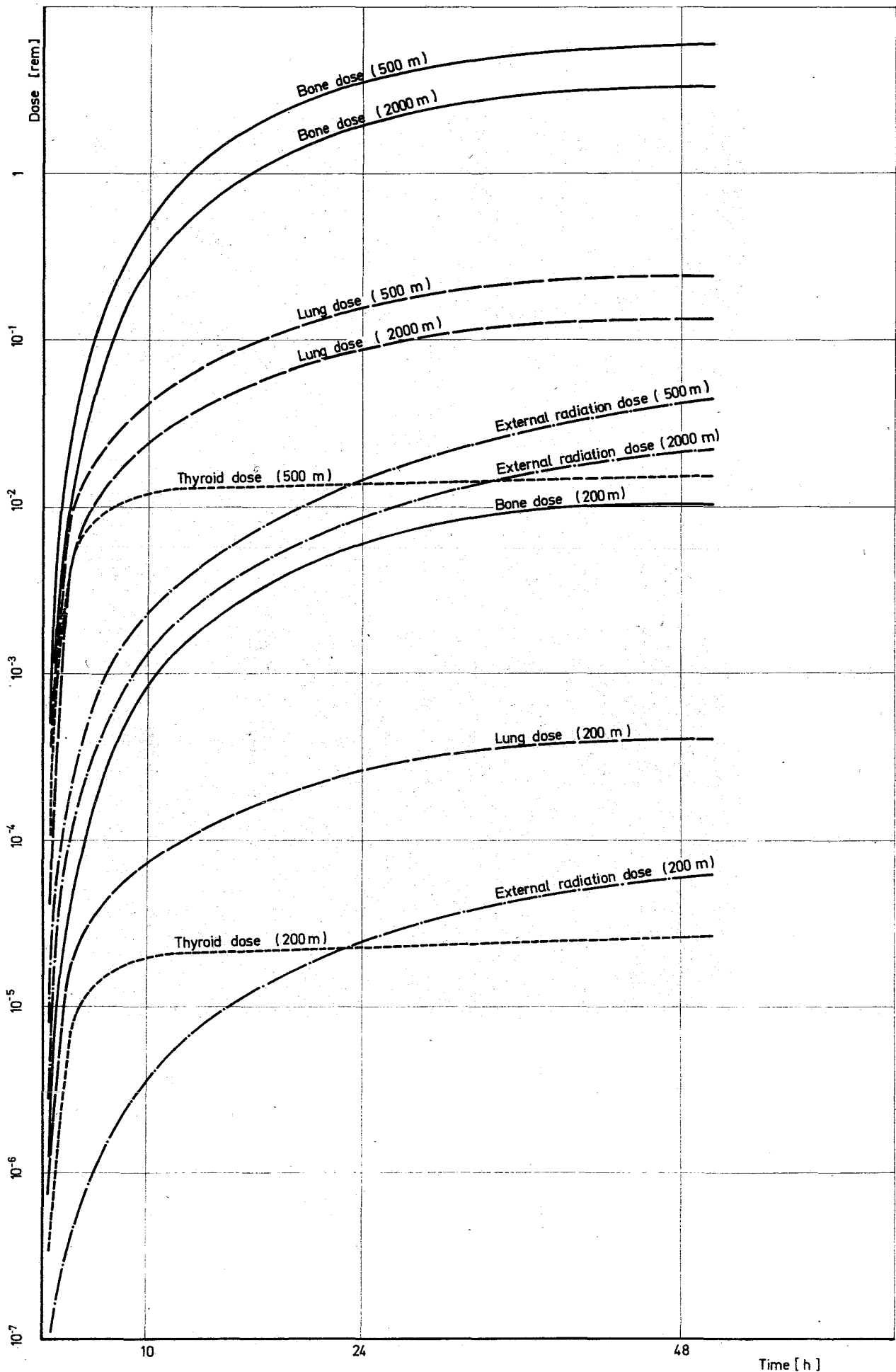


Fig.1 Large Accident Dose to different organs of typical fast sodium reactor

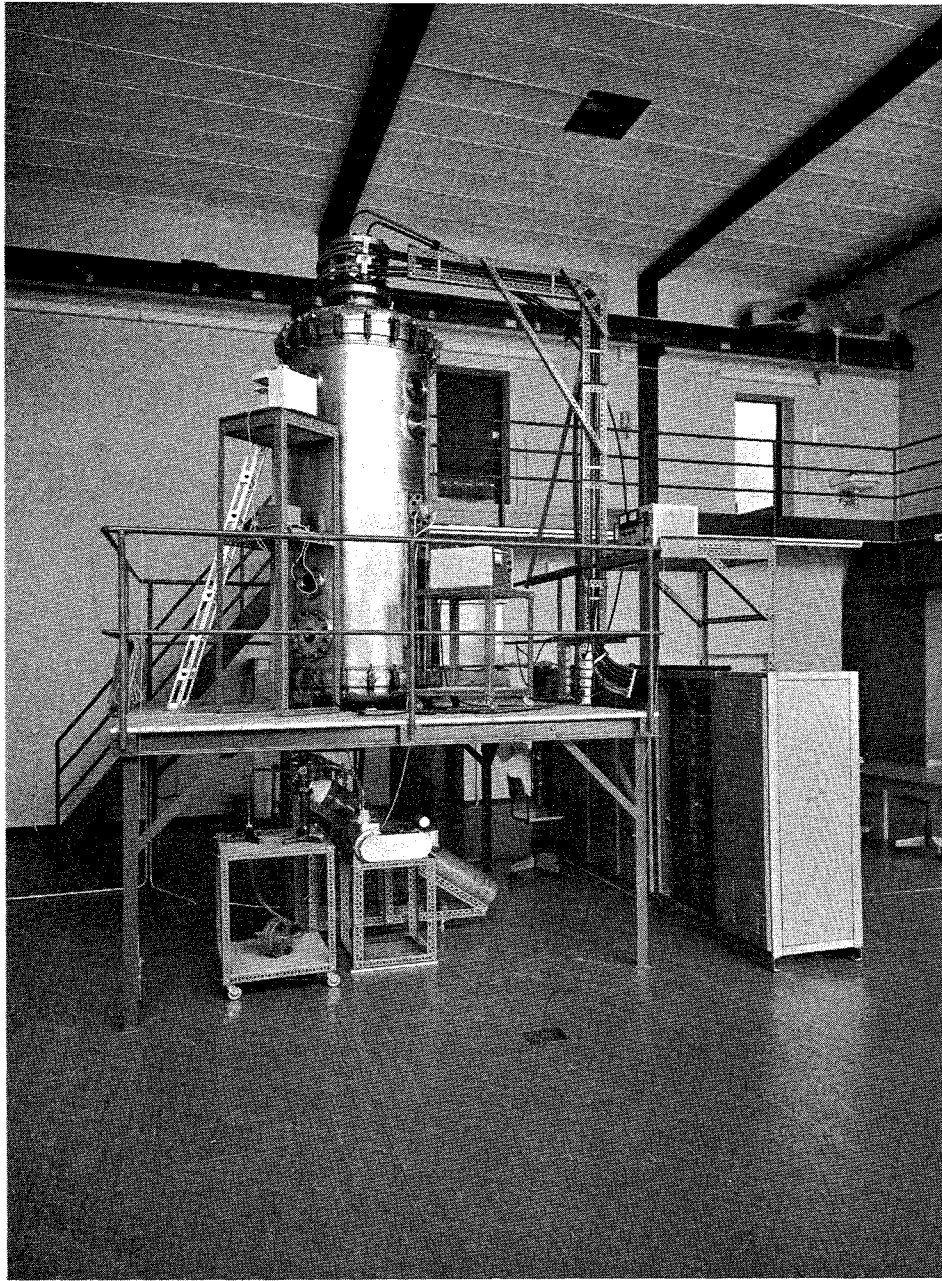
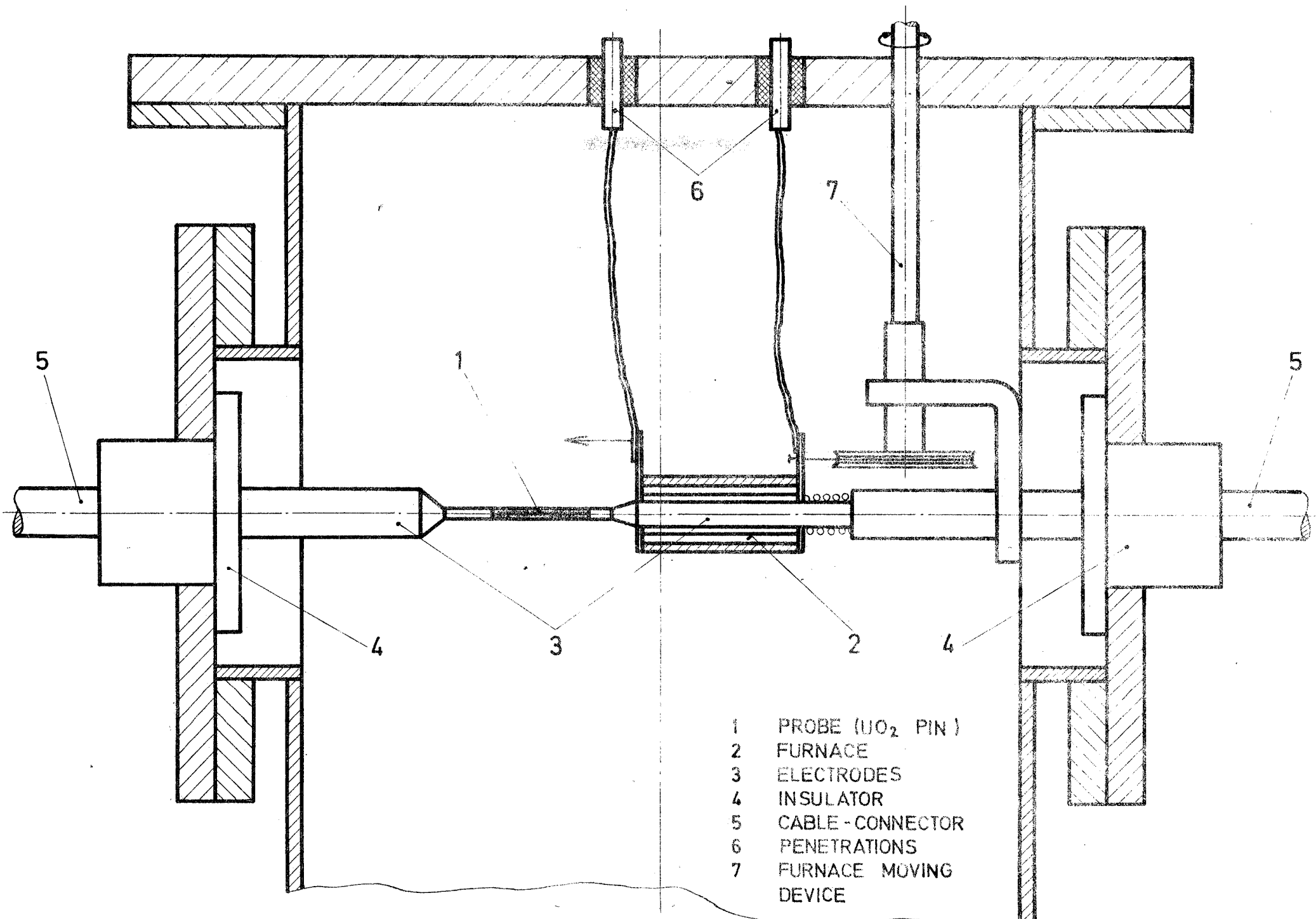


FIG. 2 TUNA



- 1 PROBE (UO₂ PIN)
- 2 FURNACE
- 3 ELECTRODES
- 4 INSULATOR
- 5 CABLE - CONNECTOR
- 6 PENETRATIONS
- 7 FURNACE MOVING DEVICE