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The Principle of Double Containment and the Behaviour of Aerosols in its Relation to the Safety of Reactors with a High Plutonium Inventory*)

by

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*) Work performed within the association in the field of fast Reactors between the European Atomic Energy Community and Gesellschaft für Kernforschung mbH., Karlsruhe

CONTENT

1. INTRODUCTION

The most important engineered safeguard against the release of radioactive materials from a nuclear power plant is the containment system consisting of containment shell(s) being sufficiently leak-tight under all circumstances including accidents credible to occur during the plant life. Because absolute leaktightness is technically not feasible, certain leakage will exist in all containment designs.

Evidently, containment systems with multiple barriers will reduce the leakage of radioactive materials released from the reactor fuel due to an accident considerably. As it has been shown earlier(1) the amount of active material released and the corresponding dose equivalent which persons at the site around the reactor plant could receive due to a large accident, can be reduced by a factor of 1000 or more if a double containment system is being applied. It should be noted that the effectiveness of a double containment system depends strongly on the course of accident chosen as a design basis for the reactor plant and the containment system which frequently is called the "Design Basis Accident". In particular the time function of pressure during the design basis accident in both the inner and outer containment governs the leakage

behaviour and subsequent release of active materials.

Therefore the radiation hazard to the environment of a nuclear power plant is in the first place influenced by the course of the design basis accident, by the leakage properties of the containment system and, of course, by the inventory of radioactive materials in the reactor. In this paper we will report on investigations done during the recent months to illuminate the interrelation between containment system, inventory of radioactive material and the radiation burden to the environment, if the activity is accidently released. Special emphasis was paid to the release of various fission products and fuel isotopes, particularly iodine and Plutonium, because of their representing the largest hazard potential to the environment of a large fast breeder reactor. The calculations were performed for the case of a typical large fast sodium cooled breeder of 1000 MWe power which is the ultimate goal of the present phase of fast breeder development.

In the following, we will (1) discuss the influence of the release parameters and the release models particularly the aerosol model on the accident doses for typical single and double containment systems, (2) we will show the significance of the decontamination of the containment atmosphere either by natural plate-out or by artificial means like filter systems, (3) draw as far as possible conclusions for the most effective containment system. Finally we will make some remarks about the most important parameters and numbers necessary to develop a reasonable description of the activity release after large accidents in fast breeders.

2. MODE OF CALCULATION

The calculations were carried out by means of the digital program MUNDO developed at Karlsruhe in the course of theses investigations which calculates the doses around a nuclear power plant due to large accidents as function of the course of accident, of the activity distribution in the containment

system following the release from the fuel, and of the meteorological dispersion in the atmosphere after leakage through the containment barriers. All the significant effects influencing the activity release as multiple containment systems, filter and air cleaning systems, plate-out behaviour, ground level or stack release can be taken into account. A block diagram given in figure 1 shows the lay-out and the capabilities of this digital program which is published elsewhere[2].

To reduce the number of parameters being important in this context the assumptions made in size and type of the power plant and in the course of accident being the cause of the activity release were kept constant in all calculations. The assumptions related to the plant were the following:

Thermal power	2500 MW
Coolant	Sodium
Load factor	0.8
In-pile time	600 days
number of fuel batches	3
Pu 239 (end of cycle)	2.13 to
Pu 240	0.61 to
Pu 241	0.07 to
Pu 242	0.03 t
Reactor building	cylindrical
Height	35 m
Radius	20 m
Stack height	75 m
Free containment volume	
primary	8000 m^3
secondary	16000 m^3
Weather conditions	
Ground level release (plane source)	Pasquill F*

* This weather condition is defined in[3].

Wind velocity	2 m/sec
Stack release (point source)	Pasquill B*
Wind velocity	2 m/sec

The assumptions concerning the course of accident were the following:

Type of accident	fast nuclear excursion
Activity release	
mechanism	melting and vaporization of the fuel
time function	instantaneous
fission gas release	100 %
other release fractions	variable
Activity transport	
distribution	homogeneous
plate-out	exponential for a limited time interval

In the calculation of doses we followed the guide lines of the ICRP-Recommendations[4]. The largest contribution to the dose values is due to incorporation. Therefore, most results discussed in chapter 3 are incorporation doses only related to the corresponding organs. Although in some cases the accident doses due to release of the noble gases can reach remarkable values we did not take them into account to avoid further complication of the results.

3. DOUBLE CONTAINMENT, RADIOACTIVE INVENTORY, RELEASE MODEL AND PLATE-OUT

3.1 The important isotopes

A number of calculations have been done to show the effectiveness of a double containment system against a single containment. Although it is easily understood that a double

* This weather condition is defined in[3].

containment reduces the radiation hazard to the public considerably the amount of reduction depends on various parameters changing with different reactor types. In thermal reactors radioactive Iodine represents the most important hazard potential because it is easily absorbed in the thyroid where it produces the thyroid dose. If we consider fast sodium-cooled breeder reactors radioactive Iodine represents likewise a major hazard potential because the Iodine built-up is proportional to the power for thermal and fast reactors. However, whereas in thermal and fast reactors the fission product inventory is approximately equal, the inventory of heavy isotopes, particularly Plutonium, in a fast reactor can be 6-8 times that of a thermal reactor with comparable power. For instance, the reference reactor considered (chapter 2) has the following inventory:

I 131	$7.9 \cdot 10^{7}$ Curie	MPC = $3 \cdot 10^{-9}$ µC/cm^3
I 132	11.1	= $8 \cdot 10^{-8}$
I 133	14.5	= $1 \cdot 10^{-8}$
I 134	15.8	= $2 \cdot 10^{-7}$
I 135	11.8	= $4 \cdot 10^{-8}$
Iodine	$7.1 \cdot 10^{8}$ Curie	
Pu 239	$1.31 \cdot 10^{5}$ Curie	MPC = $6 \cdot 10^{-13}$
Pu 240	1.31	= $6 \cdot 10^{-13}$
Pu 241	80.00	= $3 \cdot 10^{-11}$
Pu 242	0.13	= $6 \cdot 10^{-13}$
Plutonium	$8.3 \cdot 10^{6}$ Curie	

It can be derived from the maximum permissible concentrations (MPC) in air given above in the list of important isotopes that, although the Plutonium activity inventory is two decades less than the Iodine inventory, the radiological hazard potential of Plutonium exceeds that of Iodine because of the higher biological damage effectiveness of the Plutonium isotopes. This, of course, is true only for reactors

with high Plutonium inventory as fast breeder reactors are. In thermal reactors with their small conversion rate much less Plutonium builds up resulting in a higher radiological hazard potential of Iodine than of Plutonium as far as the activity inventory is concerned. Since Iodine is a thyroid seeker and Plutonium is a bone seeker the incorporation doses reported in the following tables pertain to these organs. It should be noted that the thyroid dose is produced by radioactive Iodine only, whereas in the bone dose the governing contribution comes from the Plutonium-isotopes with some smaller contribution of other fission products like Strontium and Cesium.

3.2 The release models

A number of calculations were carried out with different sets of release fractions to demonstrate the influence of assumptions made on the release of halogens and solids, particularly Iodine and Plutonium. The results are shown in table 1 in which accident doses for four different sets of release fractions as function of the leakage of the containment system are given. In the first column each line contains a set of release fractions. The first number stands for the halogens, the second for the solids, and the third for the volatile solids. Noble gases are not of interest in this context because they do not contribute to the bone or thyroid dose. Case A represents an upper limit of pessimistic assumption to our present state of knowledge. However it should be mentioned that such a set of release fractions is being discussed and considered today in various groups related to the Division of reactor licensing of the USAEC. Case B corresponds to the values of diNunno[5] taking into account a factor of 5 less for the halogens because of the good trapping capability of sodium. This set of release fractions refers for instance to the present stage of the licensing procedure for the SEFOR reactor, it also was applied in the safety analysis of the Karlsruhe Na-2 prototype reference design[6]. Case C differs from case B by assuming release fractions which correspond to the aerosol

model discussed in chapter 4. Such a set of release fractions was originally employed in the first SEFOR safety analysis but was later refused by the USAEC by reason of insufficient knowledge of the behaviour of aerosols. It should be mentioned however that the verification of this set of release fractions is the goal of the Karlsruhe aerosol program initiated in the recent months to investigate reactor fuel and fission product aerosols. Finally case D represents the aerosol model together with optimistic release fractions of gaseous and volatile fission products which could possibly be accomplished if more experimental data are available.

In table 1 the plate-out behaviour, distance from the reactor plant, and exposure time are kept constant. It should be noted that the plate-out half time of 1 h for the halogens and volatile solids and of 10 h for the solids are pessimistic. Therefore the values given in table 1 represent upper limits for the various cases. In particular the doses of case D may be decreased by a factor of 2 or more if a better plate-out is assumed and by an additional factor of 5 if the leak rate is reduced to 0.1 Vol.%/day. This means that only the accident doses of case D for the single containment system have the potential to become low enough to make a single containment design possible for large fast sodium cooled breeders.

3.3 Plate-out behaviour

Beside of the leakage characteristics of the containment system and the release models, plate-out is of similar importance which is illustrated in figure 2. Doses can be reduced by a few decades, particularly for exposures longer than 1 day, if a good plate-out behaviour of the important isotopes and their corresponding chemical compounds can be assumed. Plate-out in this context stands for both natural and artificial decontamination processes of the inner containment atmosphere. Therefore figure 2 shows also the importance of air cleaning methods like recirculation filter systems, wash-down systems or similar engineered safeguards.

From the case C (which corresponds to the case C of table 1) it can be derived that depending on the plate-out half time the thyroid dose or the bone dose is higher than the other. In figure 3 the plate-out influence for a single containment is presented showing even more the importance of this effect. Only in case C.c, i.e. the aerosol model defined in table 1 (case C) together with a good plate-out (relatively short half times) the accident doses will be in the neighbourhood of 25 rem.
This leads to the conclusion that not only the release fractions but also the plate-out characteristics of the important radioactive isotopes under the specific accident conditions should be known and therefore investigated.

3.4 Vented double containment

The double containment system considered so far in this paper consists simply of two leaktight containment shells in series. Gaseous and volatile material from this containment system is released at or close to ground level where the atmospheric dispersion is less effective than at higher altitudes. Therefore, the release of radioactive material through a stack leads to a considerable reduction of doses in the reactor vicinity. We have studied also this type of double containment system in which the outer containment volume is vented through a stack, therefore called the "vented double containment". An important parameter in the vented double containment is, of course, the circulation velocity by which the outer containment volume is vented. The blowers necessary to exhaust the air through the stack must be designed to accomplish the pressure differential between inside and outside. Therefore, depending on the in-leakage of the outer containment shell the air flow pumped through the stack will vary. In table 2 some results for a vented double containment with a 75 meter high stack are given. The important conclusion to be drawn from these numbers is the fact that for the vented double containment fairly small off-gas-flow rates through the

stack are necessary (ca. 10 Vol%/day) to achieve sufficiently low dose values, although the containment system can be improved by increasing the stackheight and using offgas filters. Again the aerosol model (case C) gives sufficiently low dose values, if we use the 25 rem bone dose as our yardstick.

4. THE RELEASE OF RADIOACTIVE MATERIALS AS AEROSOLS

From the foregoing considerations and from the results reported in chapter 3 we have learned the following:

1) The bone dose mainly produced by the release of Plutonium dominates all other doses in a large accident of a 2500 MWth fast sodium cooled breeder.

2) As long as conventional and conservative release models are applied only very leaktight double containment systems (5 Vol.%/day for the inner, and 0.5 Vol.%/day for the outer containment) or vented double containments with small stack release should be used to arrive at reasonably low accident doses.

3) If release models with low release fractions for the solids (aerosol models) can be justified, containment requirements can be relaxed considerably. Double containments with higher leakage and even single containment systems may then become possible.

The release model which we call the "aerosol model" (case C and case D in table 1 and 2 and in figure 2 and 3) is based on the following consideration: If during a large accident the fuel is melted and vaporized, essentially all the radioactive inventory contained in the fuel will be released into the containment atmosphere. The released material will then cool off rapidly. Gaseous and volatile material will stay airborne except that fraction which is plated out. Solid material, however, can stay airborne only if it is recondensed to an aerosol like smoke or dust. Certainly this will take place because recondensation from the gaseous phase is a very

effective way to produce aerosols.

The question is raised how much of the solid radioactive inventory of the fuel will form aerosols. Although this amount depends on many parameters of the fuel, of the reactor, and of the accident conditions a rough estimate can be made showing the order of magnitude which is implied: The stationary mass concentration of an aerosol is given by the following equation:

$$C = V_p \rho_p N$$

with V_p = particle volume (cm^3)

ρ_p = particle density (g/cm^3)

N = number of particles per volume (cm^{-3})

From data on smokes created in ore mines we can estimate a mass concentration of around 30 milligrams/m^3. Assuming a containment volume of 10 000 m^3, the amount of solid material staying airborne would then be 0.3 kg. This corresponds to the fraction of 0.0001 of the 3000 kg of Plutonium inventory of a 2500 MWth fast sodium cooled breeder. Although this estimate has to be proved and verified theoretically or experimentally it can be used as a first approximation demonstrating that the detailed process in the release of radioactive solid material from the fuel should be investigated to provide a more realistic picture of activity release and radiation burden to the reactor environment.

Another aspect, perhaps more important, should be emphasized. In the aerosol model we do not ask how much active material is released from the fuel (release fraction), rather we ask how much active material can stay airborne in the containment atmosphere during the accident. With other words we do not ask for release fractions 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 amount of activity released in an accident. For instance, the inner containment volume is direct proportional to the airborne aerosol mass and therefore proportional to the activity able to be released through the containment system. This means that the inner containment volume should be minimized. Effectiveness of filtering and other decontaminating systems depend as far as solid material is concerned on aerosol properties. Also deposition and inhalation behaviour of radioactive materials depend on aerosol particle size and other aerosol parameters.

We can conclude that the aerosol model is believed to provide muchmore realistic release data for solid materials and to describe much better the activity transport after large accidents in fast sodium cooled breeder reactors.

5. CONCLUSIONS

From the investigations and calculations presented the following conclusions may be drawn:

1) The presently employed release models for fission products or fuel material (case B, or in the pessimistic version case A) make it definitely impossible to employ a single containment, except one takes the position that the here considered major accident is assumed to be impossible. In that case however virtually no containment at all is necessary.
2) The extremely conservative assumptions of case A are so pessimistic that also a double containment does not give the necessary protection. The more realistic but still pessimistic case B gives satisfactory results provided that the leak rate of the inner containment is not larger then roughly 10 %.
3) In both cases, A and B, the bone dose is the limiting dose and therefore Pu instead of Iodine is the limiting factor.

4) In case of the somewhat optimistic model C which refers explicitely to the properties of Pu aerosols, the limiting factor in some cases is Iodine, in others it is the Plutonium. The function of a double containment is satisfactory even with leakage rates of 50 % in case of the inner containment.
5) Only the fairly optimistic model D making full use of (assumed) aerosol data not only for Pu but also for other isotopes gives results, which in case of a good but single containment come somewhat close to the permitted values.

   In changing from the non-vented double containment to the vented double containment it should be mentioned that the double containment system, which is vented, may not be applicable at all sites. However, if it is being applied, that means, if a stack is being provided, the results are definitely more favorable.
6) In case of a vented double containment already the realistic but still pessimistic model B gives reasonable dose rates provided that the rate of exchanging the air from the outer containment is not larger than about 100 % / day.

The over all conclusion is now, that in view of the forthcoming era of 1000 MWe fast breeder reactors there is a large and well founded incentive to put great emphasis on the investigation of aerosol behaviour in the context of fast reactor accidents.

6. REFERENCES

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(3) F.A. Gifford, "Use of Routine Meteorological Observations for Estimating Atmospheric Dispersion", Nuclear Safety 2 (4), p. 47 (1961)

(4) International Commission of Radiological Protection, "Report of Committee II on Permissible Dose for Internal Radiation" (1959)

(5) DiNunno et.al., "Calculation of Distance Factors for Power and Test Reactor Sites", USAEC-Report TID-14844 (1962)

(6) E.G. Schlechtendahl et.al., "Safety Feature of a 300 MWe Fast Sodium Cooled Breeder Reactor (Na-2)", Paper submitted to this Conference.

Table 1 : Accident doses as function of release fractions (Single and Double Containment)

500 m downwind, exposure time = 24 h
plate - out : T_H = 1 h halogens and volatile solids
T_H = 10 h solids

Leakage of Primary Containment " " Secondary " (Vol.%/day at 1 at overpressure)		Single 0.5 (∞) (Vol.%/day)	Double Containment System 5 0.5 (Vol.%/day)	 10 0.5 (Vol.%/day)	 50 0.5 (Vol.%/day)	 10 1 (Vol.%/day)
Release A 0.5 0.1 0.5	Thyroid Dose (rem)	2650	0.36	1.12	9.8	2.23
	Bone Dose (rem)	73800	147	396	3115	792
B 0.1 0.01 0.5	Thyroid Dose (rem)	530	0.072	0.22	1.96	0.44
	Bone Dose (rem)	7420	14.7	39.6	313	79.2
C 0.1 0.0001 0.5	Thyroid Dose (rem)	530	0.072	0.22	1.96	0.44
	Bone Dose (rem)	111	0.15	0.42	3.28	0.83
D 0.01 0.0001 0.1	Thyroid Dose (rem)	53	0.0072	0.022	0.197	0.044
	Bone Dose (rem)	82	0.15	0.40	3.25	0.81

Table 2 : Accident Doses for a vented double containment

500 m downwind, exposure time 24 h, stack height 75 m
Case B : plate - out T_H = 1 h halogens and volatile solids
T_H = 10h solids
Case C : plate - out T_H = 0.5 h halogens and volatile solids
T_H = 5 h solids
No filter system

Leakage of primary Containment off - gas flow secondary Containment (Vol. % of contained volume / day		0.5 1000 (Vol.%/day)	0.5 100 (Vol.%/day)	0.5 10 (Vol.%/day)
Release B 0.1 0.01 0.5	Thyroid Dose (rem)	3.4	0.5	0.05
	Bone Dose (rem)	122	41.5	5.3
C 0.1 0.0001 0.5	Thyroid Dose (rem)	1.1	0.14	0.014
	Bone Dose (rem)	0.72	0.19	0.024

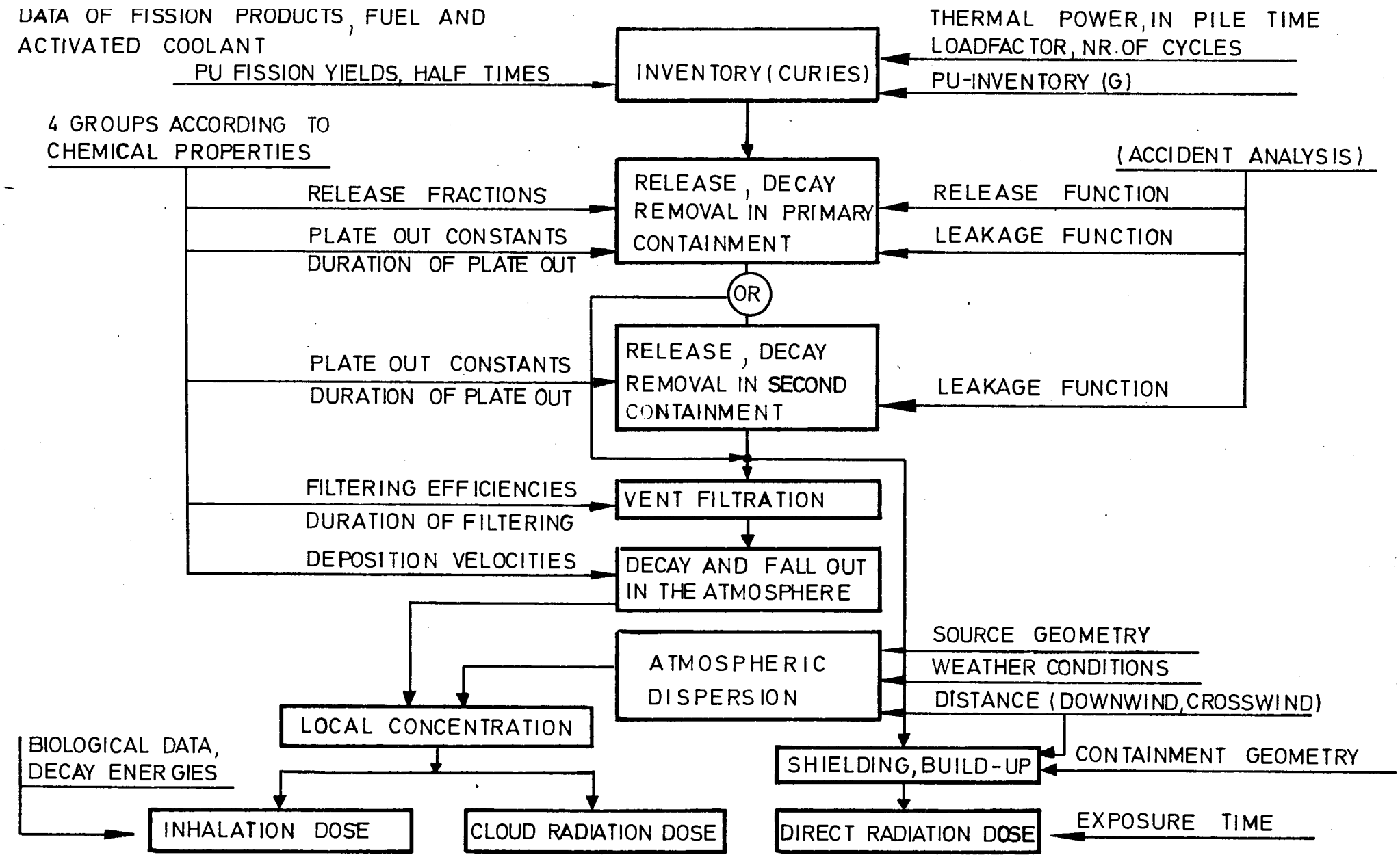

FIG.1 BLOCK DIAGRAM OF MUNDO

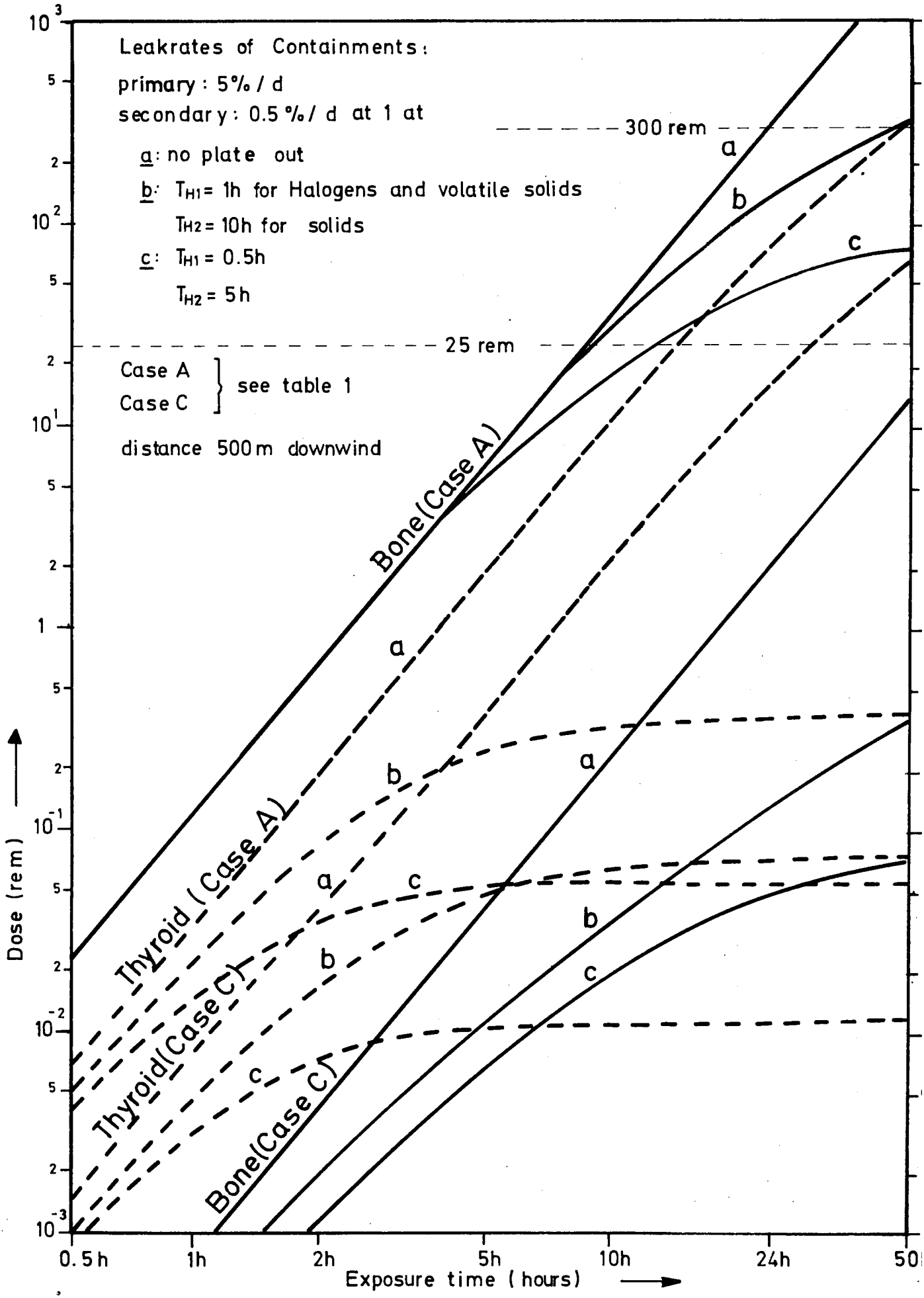

FIG. 2 Accident doses as function of exposure time and plate out (double containement)

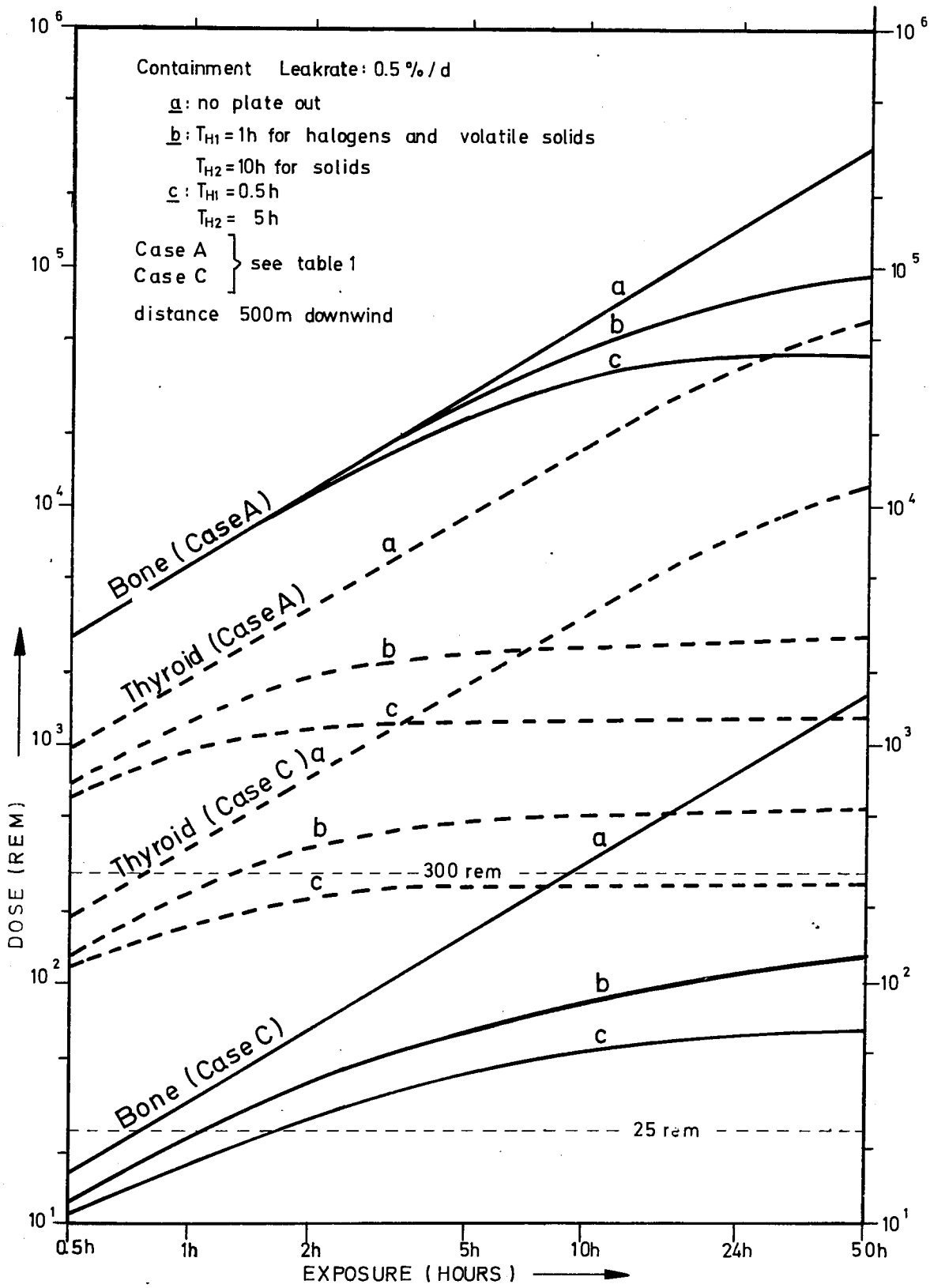

FIG.3 Accident doses as function of exposure time and plate out (single containment)