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Criticality in a spent fuel repository in wet crystalline rock

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I slutet av rapporten har bifogats en förteckning över av KBS hittills publicerade tekniska rapporter i denna serie.

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CRITICALITY IN A SPENT FUEL REPOSITORY IN WET CRYSTALLINE ROCK

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The KBS project proposes a method for final disposal of spent fuel as waste in a repository in deep mined cavities in wet crystalline rock. Such a repository would contain large amounts of fissile nuclides. This report considers the risks associated with assembly of these nuclides into critical configurations.

Criticality incidents could in theory involve plutonium as long as that remains in appreciable quantity or uranium 235 after the decay of plutonium 239, when the uranium has an enrichment making criticality in a natural water system possible.

Any criticality incident must be preceded by penetration of the long lived copper canisters surrounding the spent fuel waste, the life of which has been estimated to hundreds of thousands of years.

Hence canister failure at a time where sufficient amounts of plutonium 239 still remain is already an unlikely event. It turns out that in order to create a situation where criticality with this nuclide could take place subsequent to canister failure several other unlikely conditions have to be simultaneously assumed. Thus the overall probability of the occurence of criticality is vanishingly small.

Furthermore, it can be shown that the environmental impact of such a criticality, should it occur, will be almost negligible.

Criticality with uranium 235 involves time perspectives far longer than are otherwise considered in connection with nuclear waste disposal. Basically, it would amount to a new Oklo phenomenon, but one where the critical mass is much greater due to the lower enrichment.

For geometric and other reasons it has to take place within the mined cavities of the repository, i.e. the deposition holes for the canisters or the tunnels above them.

Criticality in the tunnels is an extremely remote possibility that could be totally eliminated by admixture of a few per cent of magnetite to the tunnel filling. The relative increase in environmental impact of the repository due to such an incident would be small.

As with plutonium, criticality in the deposition holes is a very low probability event that must be preceded by among others the partial removal of the filling (buffer material) placed in the hole. Its maximum environmental impact will be negligible.

In fact, the scenarios leading to criticality incidents in the repository are based on so unlikely assumptions that they border on physical impossibility and would hardly merit consideration in an ordinary safety analysis. The sole reason for discussing them at length in this report is the fact that statements are frequently appearing in the literature and public debate claiming that criticality represents a major hazard in geologic disposal of spent fuel.

The conclusion of this report is that, at least as far as the KBS proposal is concerned, such claims can be dismissed and that the total risk potential associated with criticality incidents in the repository can be considered negligible.

Note: Since this report was finished the KBS project decided to diminish the number of fuel rods in the canister being considered in this report from 744 to 500. The figures in table I of the report should be correspondingly reduced. The results of this change is that all reported results regarding criticality risks are overconservative.

CONTENTS

1 INTRODUCTION

A spent fuel repository contains large amounts of fissile nuclides, principally Pu 239 and U 235, sufficient for many critical masses. Hence, the possible risk contribution from criticality incidents in the repository must be assessed. This means that the probability of occurence of criticality incidents must be estimated and their environmental impact must be evaluated and compared to the overall impact of the repository.

The inventory of heavy nuclides in KBS' reference proposal is shown in table 1.

Table 1

Inventory of heavy nuclides of importance from the criticality point of view in a canister containing PWR fuel.

Time years	U 235 kg	U 238 kg	Pu 238 kg	Pu 239 kg	Pu 240 kg	Pu 242 kg
10^2	13.81	1390.2	0.10	9.37	3.41	0.65
10^3	14.04		0.	9.15	3.10	0.65
10^4	16.08			7.15	1.20	0.64
$3 \cdot 10^4$	19.14			4.08	0.14	0.62
$6 \cdot 10^4$	21.44			1.75	0.01	0.58
10^5	22.61			0.56	0.	0.54
$3 \cdot 10^5$	23.15	1390.2		0.		0.38
10^6	23.14	1390.1				0.11
10^7	22.93	1388.1				
10^8	20.99	1369.1				
10^9	8.65	1192.1				

Note: The reference repository contains about 7 000 canisters, of which the majority contains BWR fuel. The content of fissile material in these canisters is somewhat lower than the above. Other fissile nuclides (isotopes of americium, U233 and curium,are not listed since they are without importance).

Two different kinds of criticality situations can be visualized.

One is based on the separation of plutonium from most of the other constituents of the irradiated fuel and its local concentration to a region containing enough water to achieve necessary neutron moderation. As is well known the amount of plutonium needed for criticality under optimum conditions is only a few hundred grams, whereas each canister with spent fuel initially contains several kilograms of it. Hence, in principle at least, this kind of criticality incident could result from the failure of a single spent fuel canister. Obviously, this would have to happen while a significant fraction of the plutonium 239 initially available (and that produced by Am 243-decay) is still present, i.e. within the first 100 000 years or so after disposal.

The other type of criticality incident involves a critical configuration of low enriched uranium. The uranium originally present in the spent fuel will have too low an enrichment to present any criticality hazard, but in-growth from decay at Pu 239 (and Am 243) will eventually increase the fraction of the U 235-isotope to a level where a criticality incident is at least theoretically possible. However, because of the low enrichment the critical mass will be large, as shown in appendix (3) and uranium from many canisters has to join in one critical assembly (unless the filling of the mined cavities of the repository has disappeared). Thus, unlike the case with Pu 239, this type of incident can occur only a very long time after disposal.

The case of a rearrangement of the fuel with its isotopic content roughly equal to that at the time of disposal into a critical configuration can be disregarded. The critical mass is about equal to the previous case with U 235 only (i.e. the contents of several canisters) whereas the probability of occurence (e.g. the time available) is negligible in comparison.

2 REPOSITORY DESIGN OUTLINE

Although detailed descriptions of the repository design are available elsewhere, it has been considered desirable to present some key features here in order to improve the understanding of the phenomena leading to a possible criticality.

The repository is placed at a depth of about 500 m in selected, low permeability crystalline rock in a seismically quiet part of the Fennoscandian shield. The joints in these rocks are narrow, few and widely spaced (a typical condition about one joint per one to several meters and a joint width some 0.1 mm).

The essential results from the geology program carried out pertinent to the present problem can be summarized in three points:

a There are regions in Sweden where the rock is seismically and tectonically stable, i.e. any shear movements occuring are very small and limited to existing continuous weak zones. Large shear movements, that seriously upset the geometry of the repository, open up large new joints and exert major forces on waste canisters have a negligible probability.

b Joints are more or less narrow, i.e. there are nowhere volumes where a critical configuration could form (i.e. such configurations can occur only within the mined cavities of the repository).

c The flow of ground water in the joints is very slow (normally less than a liter per square meter of bulk rock cross section and year).

The spent fuel is placed in canisters that are located in holes in the bottom of horizontal tunnels, as shown in figure 1.

In the case of BWR fuel 498 full length irradiated fuel rods are placed in each canister, without any pretreatment except drying.[x)] The canister is made of 200 mm thick pure high conductivity copper. All free space between fuel rods and canister is filled with lead cast into place. Three successive lids of copper are electron beam welded to the top of the canister after filling the canister and cleaning the weld joints.

The canister is shown in a cut away section in figure 2. It is surrounded in the deposition hole with a buffer material, that fills up the volume between the canister and the rock wall as seen in figure 1. This buffer material is made up of highly compacted so called Wyoming type bentonite with a dry volumetric density in excess of 2 tonne per cubic meter. The main constituent of this material is the mineral montmorillonite in its sodium form. The tunnel above the deposition hole is filled with a mixture of quartz sand and bentonite of the above type.

When water enters the deposition holes as will slowly happen after the sealing of the repository, the bentonite will tend to absorb water and swell. If this swelling is not confined, it will continue until the material increases its volume more than tenfold.

The bentonite in the tunnel fill will take up water and swell so that most of the space between the quartz grains becomes filled with a gel with a relatively high water content. In the compacted bentonite in the deposition holes, on the other hand, very little space is available for swelling. Some expansion upward into the tunnel will take place when an equilibrium is finally obtained. In this state the compressed and somewhat displaced tunnel fill resists the pressure from the highly compacted bentonite without further movement. This pressure is a more or less permanent swelling pressure of 5-10 MPa in the bentonite. This will effectively seal off any open joints in the host rock communicating with the deposition hole and to some extent the bentonite will penetrate into these joints. However, very little of the material is lost in this way. Water filled openings cannot exist in the bentonite, since they will immediately be filled by swelling of the latter. Geologic evidence shows that the material remains stable and retains this swelling capability for millions of years.[1)]

[x)] in the case of PWR fuel the number is 744. (See however note at abstract).

The combination of this buffer material and the slow ground water flow situation in the joints of the host rock creates a very effective barrier to mass transfer. On the other hand, in order to cause appreciable corrosion of the copper canister, effective mass transfer to it of corroding substances occuring in low concentration in the ground water (such as oxygen or sulfides) is necessary, since copper does not react with water as such. The consequence is an extremely long corrosion life of the canisters, probably millions of years. An independent group of scientists has stated that "a corrosion life of hundreds of thousands of years is realistic"2). Thus the canister will outlive the plutonium 239 contained in the fuel inside it, unless premature failure has occurred due to exceptional circumstances.

3 CANISTER PENETRATION

Formation of a critical configuration must always be preceded by canister penetration. In the case of criticality involving uranium, hundreds of millions of years are available for the processes leading to a critical configuration, so that canister life is of little concern. However, in the case of plutonium criticality, it has already been pointed out that the time required for corrosion penetration of the canister is at least an order of magnitude longer than the half life of the fissile isotope. Hence, plutonium criticality need not normally be considered. Nevertheless we study early canister penetration, a very low probability event, and investigate its possible consequences such as criticality incidents and their consequences. For this reason it will be assumed that a canister penetration has occurred early in life for som unspecified reason. It could be due to som sort of fabrication defect, although it is hard to see how that could happen. It is assumed that initially a small leak develops.

One way of canister penetration that is <u>not</u> considered is mechanical demolishing due to major tectonic shear movements in the repository. Our knowledge of the geology of the area under consideration tells us that such an incident has a vanishingly small probability.

A consequence is that the buffer material will remain intact subsequent to canister penetration. Significant loss of buffer material would require major fracturing and crack opening in the surrounding rock, possible only in connection with large scale tectonic activity.

4 PROCESSES OCCURRING SUBSEQUENT TO CANISTER PENETRATION

Whereas conditions are nearly static, and fully predictable on the basis of existing knowledge as long as the canister remains intact, penetration of the latter starts a complex maze of mutually interacting processes.

These processes comprise slow corrosion of the metallic components inside the canister (lead filling and zircaloy cladding), radiolysis in the water penetrating the partly porous and fractured oxide fuel, a possible oxidation and dissolution of the fuel and various transport processes, the rate of which depend on the structure and amounts of corrosion products formed, the presence or not of gases resulting from corrosion reactions etc.

It is obvious that an accurate prediction of the real conditions as a function of time subsequent to canister penetration will not be possible.[x)] When discussing the possibility of formation of a critical configuration it will then be necessary to make conservative assumptions (i.e. in the sense of favoring criticality). When extended to all the different processes that have to be considered this eventually produces a vast difference between what can reasonably be expected to happen and what can be said with full confidence to represent conservative assumptions in every respect from the criticality safety point of view.

Thus the scenarios used below in trying to "force" a criticality will by neccessity give an impression of weird ureality.

A short review of the various processes that have to be considered is given in appendix 1.

As initially remarked in view of the long half life of U 235, considerations of the detailed processes occurring subsequent to canister penetration is of interest primarily in connection with criticality involving plutonium 239.

One basic feature that emerges from such considerations is the continuous presence of the copper canister under all conceivable conditions. It cannot be dissolved and transported away, but it may be partly converted to porous, insoluble corrosion products (such Cu_2S or Cu_2O). For this reason it is convenient to classify conceivable critical configurations involving plutonium into two categories, i.e. these occuring inside the canister and those occurring outside.

5 CRITICALITY WITH PLUTONIUM

5.1 General

From the outset it is clear that plutonium criticality requires separation of the plutonium from the bulk of the uranium and other constituents of the fuel in the canister.

As previously mentioned the critical region could be either inside the canister or outside it. Following any mechanical collapse of the canister the compressed bentonite will expand into any "free" water filled volumes remaining inside and prevent the formation of well moderated plutonium systems in them. Thus, it is only with a geometrically intact canister that inside criticality is a consideration.

It will become evident from the following that long range migration of plutonium creating a critical configuration with contributions from several different canisters will not be possible due to limitations in rate of material transport in the repository. This case is therefore not further treated.

[x)] Without extensive further experimental investigations.

5.2 Criticality inside the canister

A critical configuration inside the canister requires that uranium has been preferentially extracted (as hexavalent carbonate complex) through holes or porosities in the canister wall, while the plutonium remains behind. From the discussion in appendix 1 it is clear that this might in principle happen. Once outside the canister the uranium must have diffused away in the buffer material or alternatively been precipitated in tetravalent form by reducing agents in the latter.

Provided the canister has withstood the external swelling pressure of the bentonite the volume originally occupied by the extracted uranium has now been replaced by water. This requires that the path along which the uranium has diffused away has a sufficiently narrow porosity to prevent the bentonite from swelling and expanding into the free volume created by the disappearance of the uranium. The plutonium in oxide form is now assumed to be present in this water. The associated insoluble fission products are disregarded in the nuclear calculations.
The lead and zirconium filling must also in some way be present inside the canister, since they cannot form soluble reaction products that can be transported away. As mentioned earlier, they will both eventually corrode but with a rate difficult to establish. It turns out that if the lead filling corrodes and the oxide is converted to lead carbonate ($PbCO_3$) (as it must be if sufficient carbonate for uranium transport is available), then it alone will expand in volume enough to completely fill the void created by the removal of the uranium. No free water volume will be available and a well moderated plutonium system, a pre-requisite for criticality, cannot form.

In order to make the latter possible it is necessary to make the assumption that the lead does not corrode but remains as metal while the UO_2 is oxidized and dissolved. This appears highly unlikely but could be imagined if the cladding tubes of zircaloy remain passive and do not corrode, thus protecting the lead.

Thus, the system we are concerned with is characterized by the following:

1 Large leak in the canister (e.g. at one end)

2 Nearby leaks in the lead filling and end of zircaloy tubes

3 Lead and zirconium tubes do not otherwise corrode

4 Uranium is oxidized and extracted as uranyl carbonate complex into the buffer material. A strong horizontal "front" between extracted and untouched zones inside the fuel rods is assumed. Above this front the zircaloy tubes are filled with water containing the plutonium originally present in the uranium that has disappeared.

For any given time after disposal and amount of uranium extracted, the length of water filled cladding tubes and the amount and isotopic composition of plutonium contained therein (as a dispersion of insoluble oxidic material) are given. The situation is depicted in figure 3.

Reactor physics calculations have been carried out using the ASEA-ATOM multigroup program MIC0 (appendix 2). The infinite neutron multiplication factor was calculated assuming a lattice of tubes filled with plutonium-oxide slurry in water, in which the water content was varied from zero to 99 %. The calculations were made for different times, each time corresponding to a given isotopic composition of the plutonium.

In this way it was found that k_∞ exceeded unity for most water ratios considered. However, in order for a critical configuration to be possible, the neutron leakage must be sufficiently low.

The neutron leakage is determined by the material composition of the assembly, its dimensions (in this case diameter and height, where the diameter is now always equal to the inner diameter of the canister) and by the reflector saving. The latter was separately calculated.

With the radius given the height required for criticality can now be calculated as a function of age (i.e. isotopic composition).

The figures are shown in column 4 of table I, appendix 2.

For any given time the maximum amount of uranium that has left the canister is limited by the rate of transport processes, however. The calculated figures are given in appendix 1, section 7. These amounts can be translated into an equivalent height of the UO_2 column that has been replaced by water, given in column 5 of the table. It is seen that the latter is at all times much smaller than the height required for criticality i.e. criticality will never be possible.

It should be observed that the nuclear calculations are based on the peculiar assumption that the insoluble plutonium oxide is homogenously dispersed in an optimum fraction of the available water volume, whereas in reality it would tend to settle on the bottom. This would strongly decrease the reactivity.

Thus it can be definitely concluded that inside criticality is impossible as long as the bentonite buffer material remains in place.

As discussed in section 5.3 losses of a major fraction of the bentonite (which is not considered feasible on the basis of geologic evidence) would largely eliminate the impediment to uranium transport in the deposition hole outside the canister. If furthermore, due to the difficulties in modeling evident from the discussion in appendix 1, the transport limitations inside the canister are neglected, the loss of uranium could occur considerably more rapidly. Under such conditions the theoretical potential for inside criticality could arise.

Below a list has been compiled of the large number of unlikely assumptions that have to be combined in order to make a criticality possible, compared in each case with what is considered a realistic assumption.

This list should be sufficient evidence that the scenario leading to criticality discussed above, can be dismissed as utterly unrealistic.

	Realistic assumption	Conservative assumption employed to achieve a criticality
1. Canister integrity	Canisters will not be penetrated until practically all Pu 239 has decayed(after a million years or more.) Probability of initial fabrication defect extremely low. (Penetration is nevertheless assumed in this column below).	Canister is penetrated at time zero.
2. Copper corrosion subsequent to penetration	No appreciable further corrosion. Hole in copper will be plugged with lead corrosion products and little more will happen.	Extensive corrosion will occur transforming at least upper part of canister into porous copper oxide that offers little diffusional resistance. Corrosion caused by O_2 and H_2O_2 formed by radiolysis in fuel zone.
3. Lead corrosion (initial)	Some intial corrosion will occur, plugging hole in copper. Thereafter little will happen.	Lead will corrode so that water gets into contact with all fuel rods e.g. at the top end. The lead is transformed into a porous crust of corrosion products offering little diffusional resistance.
4. Lead corrosion (long term)	As in (3). If against expectation extension initial corrosion has actually occured, corrosion will continue in the longer term coverting the lead to e.g. carbonate. Accompanying volume expansion will fill any voids inside canister.	Lead corrosion stops following initial phase in which contact with all fuel columns is achieved. The lead metal then remains inert, i.e. voids created by uranium disappearance are not filled with lead corrosion product.

	Realistic assumption	Conservative assumption
5. Zircaloy corrosion	The zircaloy takes perhaps tens of thousands of years to corrode into oxide and effectively prevents uranium and and plutonium migration during much of this time. However, it will eventually end up as zirconium-dioxide.	The zircaloy tubes (e.g. the upper ends) are penetrated by corrosion to create a pathway for uranium migration. Thereafter corrosion largely ceases, so that the zirc.tubes remain intact for the time under consideration.
6. Water radiolysis	Very little net radiolysis occurs. There is little water volume in contact with the fuel. The presence of excess hydrogen (from Zr-corrosion) reduces any fuel or copper metal oxidized due to radiolysis.	Considerable crack volume in fuel causes rapid radiolysis due to alpha radiation. Hydrogen produced remains inert and leaves as gas. Oxygen and H_2O_2 cause oxidation reactions.
7. Fuel - water reactions	Very little net reaction. Fuel largely unaffected, negligible out-transport of hexavalent carbonate complexes (cf Oklo experience) due to lack of oxidation and extremely narrow passages out of canister.	Uranium oxidized to hexavalent stage by O_2 and H_2O_2. H_2 chemically inert and leaves as gas. UO_2 lattice transformed into higher oxide. Fission products tied up in lattice are able to leave. Uranium removed as hexavalent carbonate complex through porous corrosion product layers, the latter providing little assistance to transport. Plutonium remains behind as oxide slurry.
8. Conditions in the buffer material	Conditions largely unchanged. Very slow zircaloy corrosion causes hydrogen production. Gas (H_2) bubble will eventually appear, but probably only long after the plutonium has decayed.	Most of the bentonite lost through large new cracks that have opened (an impossibility according to geologic expertise.) Gas bubble may obstruct transport processes but this is neglected.

	Realistic assumption	Conservative assumption
9. Plutonium location	In case a free water volume would actually exist any plutonium oxide present would settle on the bottom, where heavy self shielding would prevent criticality.	Plutonium oxide is assumed to be homogenously suspended in an optimum free water volume, although no known mechanism for achieving this is available.
10. Insoluble fission products	Insoluble fission products (such as rare earths) remain behind and reduce the reactivity with 5-10 %.	Insoluble fission products are extracted with the uranium.

5.3 Criticality outside the canister

A plutonium criticality outside the canister requires a dissolution and subsequent reprecipitation of the plutonium.

Selective dissolution of plutonium from the fuel matrix, leaving the uranium behind, is not considered possible. First of all, the conversion to the hexavalent state, which makes possible the formation of soluble complexes, occurs preferentially with uranium. (This is what creates the theoretical potential for Pu criticality inside the canister). Plutonium is much harder to convert to hexavalent and is unstable in that valence stage. Secondly, no complexing agents specific to plutonium are available either in the ground water or the buffer material. (So called fulvic acids that are available in the ground water are not specific in this sense). The plutonium will either be extracted in parallel with the associated quantity of uranium or remain behind.

The former alternative creates a theoretical potential for criticality provided plutonium is selectively reprecipitated after the uranium-plutonium mixture has diffused out of the canister. If the plutonium arrives into the buffer material zone in the hexavalent stage (produced by oxidating radiolysis effects) this could indeed be the case.

Criticality calculations have been made assuming a bentonite-PuO_2-mixture of spherical geometry (appendix 2). The water saturated bentonite is assumed to have a bulk density of 1.95 and a water content of 29.6 % 10) before PuO_2 precipitation. The PuO_2 is assumed to be free of fission products. The situation is depicted in figure 4. The results are summarized in table 3.

Table 3 Minimum critical masses of Pu in bentonite

Time	0	10^4	$3\cdot10^4$	$6\cdot10^4$
Min crit. mass kg Pu fiss	5.3	3.2	2.3	2.2
Plutonium conc correspond to crit. mass kg/dm^3	0.05	0.05	0.04	0.04
Total quantity of Pu (fiss) available (remaining inside canister and migrated to outside)	8.9	6.7	3.8	1.6

It is seen that on outside criticality must occur within about 50 000 years after disposal. Beyond that time not even the whole inventory of fissile plutonium can form a critical configuration.

However, as shown in appendix 1, section 7, transport limitations determine the maximum amount of uranium that can have left the deposition hole at a given time. The figures are given in table I in appendix 1.

The maximum amount of "separated" fissile plutonium that could be available in the buffer material is equal to the fraction of the total inventory of uranium that has left the deposition hole times the total inventory of fissile plutonium. A comparison of these quantities with the plutonium critical mass is given in table 4.

Table 4 Minimum critical mass of Pu vs max. amount available in bentonite

Time y	Min crit mass kg Pu fiss	Max amount available in buffer material kg Pu fiss
0	5.3	1.0
10^4	3.2	1.0
$3\cdot10^4$	2.3	0.9

It is seen that the amount available is several times less than that required even assuming that it collects into a perfectly spherical volume completely free of uranium. (The latter is of course a preposterous proposal). Thus plutonium criticality in the buffer material outside the canister can be confidently disregarded as long as the bentonite remains in the buffer zone.

If the bentonite largely disappears (as mentioned elsewhere, there is no reasonable physical way in which this could happen) and is replaced by water, the critical mass of plutonium will decrease because of the disappearance of the diluting and absorbing action of the bentonite minerals. At the same time the diffusivity will increase, making possible a higher rate of removal of uranium into tunnel and rock joints.

The calculated critical masses as a function of percentage remaining bentonite are shown in table 2 of appendix 2.

The "available" amounts of plutonium will now be higher than those shown in the right column of table 4, perhaps by a factor of 2-4. (An accurate estimate is not possible since the disappearance of much of the bentonite and the corresponding increase in diffusivity, may be associated with changes in geometry that cannot be defined). Comparing the figures so obtained with the critical masses given in appendix 2, the conclusion evidently must be that criticality cannot be entirely ruled out under these extremely unlikely conditions. However, attention should be called to the fact that all transport resistances inside the canister have been neglected. Any realistic modeling of these resistances, such as proposed in ref (13), would probably show that the risk of criticality is not real. However, due to the complexities mentioned in appendix 1, such modeling was not attempted.

As in section 5.2 a list has been compiled of all the unlikely conditions that have to be met to create a criticality situation as described above. As can be seen, the assumptions are quite different in some cases.

These assumptions are contrasted below with those considered representing realistic conditions. The contrast should illustrate how remote a possibility criticality really is.

	Realistic assumption	Conservative assumption
1. Canister integrity	Canisters will not be penetrated until practically all Pu 239 has decayed (a million years or more). Probability of initial fabrication defect extremely low.	Canister is penetrated at time zero.
2. Copper corrosion subsequent to penetration	No appreciable further corrosion. Hole in copper will be plugged with lead corrosion products and little more will happen.	Extensive corrosion will occur transforming at least upper part of canister into porous copper oxide that offers little diffusional resistance. Corrosion caused by e.g. O_2 and H_2O_2 formed by radiolysis in fuel zone.
3. Lead corrosion	Some initial corrosion will occur, plugging hole in copper. Thereafter little will happen.	Lead will corrode so that water gets into contact with all fuel rods. The lead is transformed into a porous crust of corrosion products offering little diffusional resistance.

	Realistic assumption	Conservative assumption
4. Zircaloy	The zircaloy takes perhaps tens of thousands of years to corrode into oxide and effectively prevents uranium and plutonium migration during much of this time.	Zircaloy reacts with water and is transformed to porous ZrO_2 in a time short compared to the half-life of Pu 239.
5. Water radiolysis	Very little net radiolysis occurs. Only a small water volume is in contact with the fuel. The presence of excess hydrogen (from Zr corrosion) reduces any fuel or copper metal oxidized due to radiolysis.	Considerable crack volume in fuel causes rapid radiolysis (due to alpha radiation). Hydrogen produced remains inert and leaves as gas. Oxygen and H_2O_2 cause oxidation reactions.
6. Fuel - water reactions	Very little net reaction. Fuel largely unaffected, negligible out-transport of hexavalent carbonate complexes (cf Oklo experience) due to lack of oxidation and extremely narrow passages out of canister.	Fuel (U + Pu) oxidized to hexavalent stage by O_2 and H_2O_2. H_2 chemically inert and leaves as gas. UO_2 lattice transformed into higher oxide. Fission products tied up in lattice are able to leave. U and Pu removed as hexavalent carbonate complexes through porous corrosion product layers, the latter providing little restance to transport.
7. Conditions	Conditions largely unchanged. Very slow zircaloy corrosion causes hydrogen production. Gas (H_2) bubble may eventually appear, but probably only long after the plutonium has decayed.	Most of the bentonite lost through large new cracks that have opened. Gas bubble may obstruct transport processes but this is neglected.

	Realistic assumption	Conservative assumption
8. Plutonium chemistry	Plutonium remains in the tetravalent stage and is not transported by carbonate to a significant extent even in case water contacts the fuel. There is very strong experimental evidence that this is the case in irradiated fuel leaching under oxidizing conditions.	Plutonium is radiolytically oxidized to the hexavalent stage, in spite of the presence of free hydrogen etc, transported away as carbonate complex through lead and copper corrosion products and later selectively reprecipitated as tetravalent in the bentonite.
9. Plutonium migration	No significant transport of plutonium into bentonite zone. Small amounts transported precipitated in irregularly shaped region.	All plutonium leaving the canister concentrated into a spherical with optimum diameter volume, essentially free of uranium.

5.4 Consequences of a criticality

A plutonium criticality leading to significant energy release will produce radiotoxic nuclides in addition to those originating from the initial inventory of the failed canister.

In order for the criticality phenomenon to be important in this respect those radionuclides critical with regard to environmental effects have to be produced in an amount of the same order of magnitude as the preexisting inventory in the canister. However, calculations show that this is not the case, so that the direct radiologic aspect of the criticality incident is unimportant (appendix 2).

A more crucial question is the effect of the criticality on surrounding intact canisters. If these were affected in such a way that failure is induced, the criticality could spread like a disease in the repository and cause large scale failure of the canisters.

The effect on adjacent canisters must be via their chemical environment or their temperature.

A neutron chain reaction may produce some oxidizing radiolysis products, such as free oxygen. In order to reach adjacent canisters this oxygen has to migrate in the rock joints, since it would be reduced if diffused into the tunnel[x]. Calculations show that some temporary increase in the corrosion rate of adjacent canisters could result if the flow is directly from one deposition hole to the next. However, even in this case the corrosion that could take place before the "reactor" has shut down due to plutonium decay and burn up would be completely insufficient to cause penetration (appendix 4).

*) Initially it will be reduced in the rock joints near the hole too, but it is assumed that the amounts of oxygen involved are large enough to exhaust the inventory of ferrous minerals there that cause the reduction.

The maximum temperature in a critical region will be equal to the saturation temperature at the hydrostatic pressure at repository depth (about 5 MPa), i.e. $\sim 265^{o}C.^{x)}$ It can then be assumed that this is also the temperature at the surface of the whole copper canister, because of the high thermal conductivity of the copper. Calculations have been performed (appendix 5) to find out how this affects the temperature of adjacent canisters. The results show that the resulting temperature increase is limited (max $45^{o}C$). Such a temperature rise will not significantly decrease the corrosion life of the canister since it will last only for a small fraction of their predicted lifetime. Burning of the available plutonium will be able to sustain such an overtemperature for a time of about 300 years only.

Thus local criticality in a canister that has failed prematurely due to a fabrication defect or other unknown reasons will not have any important consequences for the surrounding canisters in the repository.

6 CRITICALITY INVOLVING URANIUM 235

After the complete decay of Pu 239 and Am 243 the remaining uranium has a maximum enrichment of 1.66 %.

With this enrichment and the nuclide inventories given in table I there is a possibility of criticality in a system moderated with natural water. Such a criticality could a priori take place in either of the following locations:

- The deposition holes
- The tunnels and shafts of the repository
- The joints in the host rock in which the repository is placed.
- Regions with porous material, such as soil, away from the repository to which the uranium has been transported by the ground water.

Elementary criticality calculations and general geochemical and hydrological knowledge make possible the following conclusions concerning the latter two alternatives.

- The dimensions of the joint system in the host rock is such that criticality is excluded for geometry reasons alone (too much neutron leakage, i.e. the geometry is "eversafe").
- Criticality in a soil region away from the repository requires long range transport of uranium. This will not be possible due to the insolubility of the tetravalent uranium that exists under the reducing conditions in deep ground water. (Long range transport of uranium resulting in ore formation occurs under oxidizing conditions).

Thus we are only concerned with criticality incidents in the mined cavities of the repository.

[x)] A higher temperature would lead to water vaporization, drying out of the "reactor" and shut down of the chain reaction.

6.1 Probability of occurence of criticality

6.1.1 Criticality in the tunnels above the deposition holes

Criticality calculations assuming presently prescribed composition and water content of the quartz-bentonite mixture in the tunnels have been made. The result show that the minimum critical mass is 4 400 kg of uranium assuming spherical geometry (appendix 3). This means that uranium from several (at least 4) canisters has to participate in the critical assembly, requiring long range migration along the tunnel and local precipitation in a small region.

Such processes are wellknown in uranium ore genesis. In this case the uranium is transferred to the hexavalent, relatively soluble form in mineral wheathering under aerated conditions. When this ground water containing the uranium reaches a reducing region (either because of oxygen consumption due to its own content of organic matter or by admixture with a water flow of different composition) reduction to the almost completely insoluble tetravalent form occurs.

In order for a critical configuration to be formed, this process has to occur in the tunnels of the repository. Migration as such in either the hexavalent soluble form (which could occur with a significant rate) or in the nearly insoluble tetravalent state (which would be immeasurably slow) would not produce a local concentration of the uranium.

The ground water chemistry at depth will reduce the uranium to the tetravalent state, thereby immobilizing it and preventing any further local concentration. Thus the transport leading to a concentration must occur in the hexavalent state, as in ore genesis, and must take place before chemical equilibration with the regional ground water flow has occurred.

Initially, presence of high concentrations of alpha-emitting nuclides may create oxidizing conditions that counteract the reducing effect of the ground water, perhaps enabling the uranium to remain in the hexavalent form. However, after a few million years nearly all of this is gone and we are basically dealing with a uranium ore (with an isotopic composition somewhat deviating from normal). Under these conditions precipitation in the tetravalent form due to reduction by the ground water must occur. Using an hydraulic gradient of 1 %, a permeability of 10^{-9} m/s (established for the tunnel filling) and the measured value of chromotographic retention factor it can be shown that any hexavalent uranium will be transported in the tunnel only about 10 meter in 10 million years. The average diffusion distance is of the same order. This shows that migration along a tunnel is not a reasonable mechanism for creating a criticality.

In principle accumulation of a critical mass in the tunnel could occur due to some kind of "focused" uranium bearing flow in rock joints that communicate with many canisters. If the the reducing capacity of the minerals in the walls of these joints become exhausted, precipitation in the tunnel could occur due to its higher reducing capacity. It goes without saying that such a scenario is too unrealistic to deserve attention.

Furthermore, by adding about 7 % of magnetite to the quartz sand used, criticality calculations show that the neutron absorption capacity of the tunnel filling can be made high enough so that criticality can never occur with any amount of uranium.

There is no way in which the largely coarse grained insoluble quartz and magnetite could be dissolved or dispersed and removed from the tunnel by the ground water flow.

Thus, it can be concluded that the probability of the occurrence of criticality incidents in the tunnels above the deposition holes is negligibly low and could if desired be altoghether eliminated by a slight change in the composition of the tunnel filling. This change is inexpensive and of no consequence for the function of the repository in other respects.

6.1.2 Criticality in the deposition holes

As previously mentioned criticality in the tunnel filling requires the presence of the uranium from several canisters. Because of the somewhat higher neutron absorption capacity of the bentonite compared to quartz sand, the quantity required for criticality in the deposition holes is even higher.

It goes without saying that the transport of uranium away from <u>several</u> deposition holes and into <u>one</u> other such hole is so strange a scenario as to hardly merit consideration. Furthermore, if any uranium diffusing into the tunnels is effectively anchored there by reduction to the tetravalent stage, the tunnels may be considered eliminated as transport avenues between the deposition holes. There is no way in which uranium transport through the joints in the host rock from one deposition hole to another could cause the required accumulation in the latter. Thus it can be concluded that as long as the bentonite remains in the deposition holes, there is no way in which a critical configuration could be formed in one of them.

As has previously been pointed out, on the basis of present knowledge of the conditions at 500 m depth in selected stable rock away from weakness zones, no mechanism can be conceived by means of which a major fraction of the bentonite could leave the deposition hole. This would require the opening up of very large cracks into which the bentonite gel would expand. Nevertheless criticality calculations have been carried out assuming that this has happened.

These calculations show that criticality could occur with the uranium contained in a single canister if ∿45 % of the bentonite originally present in the deposition hole has been lost. Such a criticality must be based on a type of scenario like the following:

1) Loss of bentonite due to large crack openings in the rock

2) Penetration of the canister.

3) Diffusion of uranium (hexavalent) into the upper part of the deposition hole.

4) Diffusion of ferrous iron down from the tunnel.

5. Precipitation of the uranium in the tetravalent form in the high-water-content bentonite.

6.1.3 Over-all probability of criticality

From the above it can be seen that criticality will not be possible in the mined cavities of the repository as long as the tunnel filling on bentonite buffer material remain in place (at least if som magnetite is added to the tunnel filling).

Disappearance of the tunnel filling is physically impossible. Disappearance of a major fraction of the bentonite buffer material in the deposition holes is incompatible with present ideas of the behaviour of rock formations of interest for location of a repository, and so is an exceedingly low probability event. Criticality is a low probability event in a deposition hole where this has happened.

6.2 Consequences of criticality

The attainment of a critical configuration will be a slow process resulting from gradual accretion of material. Criticality as such will be of no special significance since the reactor will have strongly negative temperature and power coefficient. No sudden energy release is possible. In order for energy to be released the temperature must rise and this requires the addition of more uranium to keep the assembly critical. The maximum temperature that can be reached in the critical region is the saturation temperature at the hydrostatic pressure in the repository. A temperature exceeding that would evaporate the water from the critical region and make it subcritical.

6.2.1 Criticality in tunnels

As has been shown any risk of criticality in the tunnels can be eliminated. Nevertheless, calculations reported in appendix 3 were carried out of the maximum consequences of such a criticality, assuming that the uranium from all the holes in one tunnel participate in the critical configuration.

Appendix 3 considers the maximum generation of power (and toxic radionuclides) that is theoretically possible from all the uranium disposed in the bottom holes of one tunnel, while disregarding the negligible probability that such a configuration is ever realized.

It is shown that the power of such a "reactor" must be less than 130 kW.

An upper limit of the total energy generated by such a reactor before it shuts itself down due to depletion of fissile material will be some $3 \cdot 10^6$ MWd, i.e. equal to the burn up in some 900 tons of disposed power reactor fuel.

Since such an energy generation would be distributed over at least a period of some 100 000 years, the release of radionuclides to the ground water would correspond to less than one failed canister in the repository every 1000 years. This is an order of magnitude lower release than has been assumed in assessing the over-all radiologic impact of the repository. Consequently, the radiologic impact of this kind of criticality will be small in comparison with the latter, which are given in the KBS main report on spent fuel disposal. (Short lived radionuclides such as Sr 90 and Cs 137 will always decay before reaching the biosphere).

6.2.2 Criticality in deposition holes

As previously mentioned the discussion can be limited to incidents involving the uranium contents of a single canister, possible only after the loss of most of the bentonite buffer material. The amount of fission products and radiotoxic heavy nuclides created by such a criticality is strictly limited and can be calculated to be only ∿40% of the initial inventory of such nuclides in the canister. This means that the environmental consequences are only a corresponding fraction of those resulting from an initial failure of a freshly disposed canister followed by rapid extraction of the fuel into the ground water. (These will be given in the KBS main report on spent fuel disposal).

7 SUMMARY AND CONCLUSIONS

A spent fuel repository contains large amounts of fissile nuclides, principally Pu 239 and U 235. This report considers the probability and consequences of the formation of critical configurations in the repository due to their presence.

Such critical configurations could be of two kinds. Criticality could be achieved by separating plutonium from the bulk of uranium and concentrating it locally. This would obviously have to occur before most of the Pu has decayed to uranium 235, i.e. within roughly the first 100 000 years after disposal. It is sufficient to consider the plutonium content of a single canister in this context.

The other kind of criticality involves enriched uranium. After the plutonium has decayed to U 235 the enrichment of the uranium will be high enough to make criticality theoretically possible in a natural water moderated system. Because of the long life of U 235 this possibility exists for periods of time much longer than are otherwise considered in connection with disposal of nuclear waste. These two types of criticality incidents are separately considered.

Because of the slow rate of transport of materials in the repository criticality with plutonium must occur in the vicinity of the canister from which it originates.

Any criticality phenomenon in a spent fuel repository will develop slowly, occurring as a result of gradual accretion of material deposited in the critical region by flowing ground water or by diffusion. Following attainment of criticality, power generation will build up very slowly as a result of further fissile material addition, due to the negative temperature coefficient of reactivity. Sudden, explosive energy release is physically impossible.

Normally the canister will outlive the plutonium 239 by a wide margin, so that canister penetration necessitating criticality considerations will be an exceptional case (due e.g. to faulty fabrication).

For analyzing the probability of plutonium criticality it is necessary to discuss the complex interacting corrosion and solution processes initiated by penetration. A short review of these processes is given in appendix 1.

Because of considerable uncertainties it is necessary to make many conservative assumptions favoring the formation of a critical assembly. These in combination will create a model of the system which tends to be more or less far removed from reality in a way favoring the occurence of criticality.

In all conceivable models involving plutonium criticality the canister remains as a component of the system, although it may be corroded and penetrated to various degrees. For that reason it is convenient to consider criticality events inside and outside the canister separately.

Criticality inside the canister could result if the uranium is selectively extracted as soluble complex and the plutonium remains behind in the water volume created by the disappearance of the uranium. However, even disregarding material transport limitations inside the canister, it can be shown that sufficient quantities of uranium cannot be extracted and removed from the canister and bentonite buffer in time to make possible accumulation of a critical mass of plutonium. This kind of criticality is possible only if most of the bentonite is lost through large cracks appearing in the rock around the deposition hole. This is considered impossible in view or existing knowledge of the behaviour of the type of rock to be used for the repository.

Criticality outside the canister requires that uranium and plutonium are extracted together and the plutonium then selectively precipitated in the bentonite buffer outside the canister. (Selective extraction of plutonium without uranium is not possible). As in the previous case it can be shown that such a criticality is possible only if the bentonite buffer material is largely lost in some inexplicable way.

The consequences of criticality with plutonium are in any case relatively unimportant. The amounts of long lived radiotoxic nuclides created is small in comparison with the already existing inventory in the canister. The possible temperature increase or local change in the ground water chemistry occurring will not significantly affect neighboring canisters in the sense of increasing the risk of their penetration.

For criticality geometry reasons criticality with uranium could occur only within the mined cavities of the repository.

Criticality in the tunnels above the deposition holes is considered a very remote possibility since no processes can be envisaged whereby the necessary local concentration could be achieved.

By increasing the neutron absorption capacity of the tunnel filling by addition of a few per cent of magnetite criticality in it can be entirely ruled out.

The upper theoretical limit of the amount of long lived fission products and radionuclides formed due to such a criticality is about 10 % of the whole original repository inventory, appendix 3. The radiological impact is well below that estimated for the repository under normal conditions.

Uranium criticality in the deposition holes can occur only if most of the bentonite has been lost through joints in the host rock - which as mentioned is considered impossible.

The consequences of such a criticality are limited due to the limited amount of uranium involved and correspond roughly to the release of ∿40 % of the original nuclide inventory of one canister into the ground water at the repository location.

Summarizing, the probability of criticality incidents in the repository is exceedingly small. The maximum radiologic consequence of criticality incidents is small enough to stay below existing radiation hazards on the earth surface by a wide margin. The conclusion, then, is that criticality incidents in the repository represent a negligible risk potential.

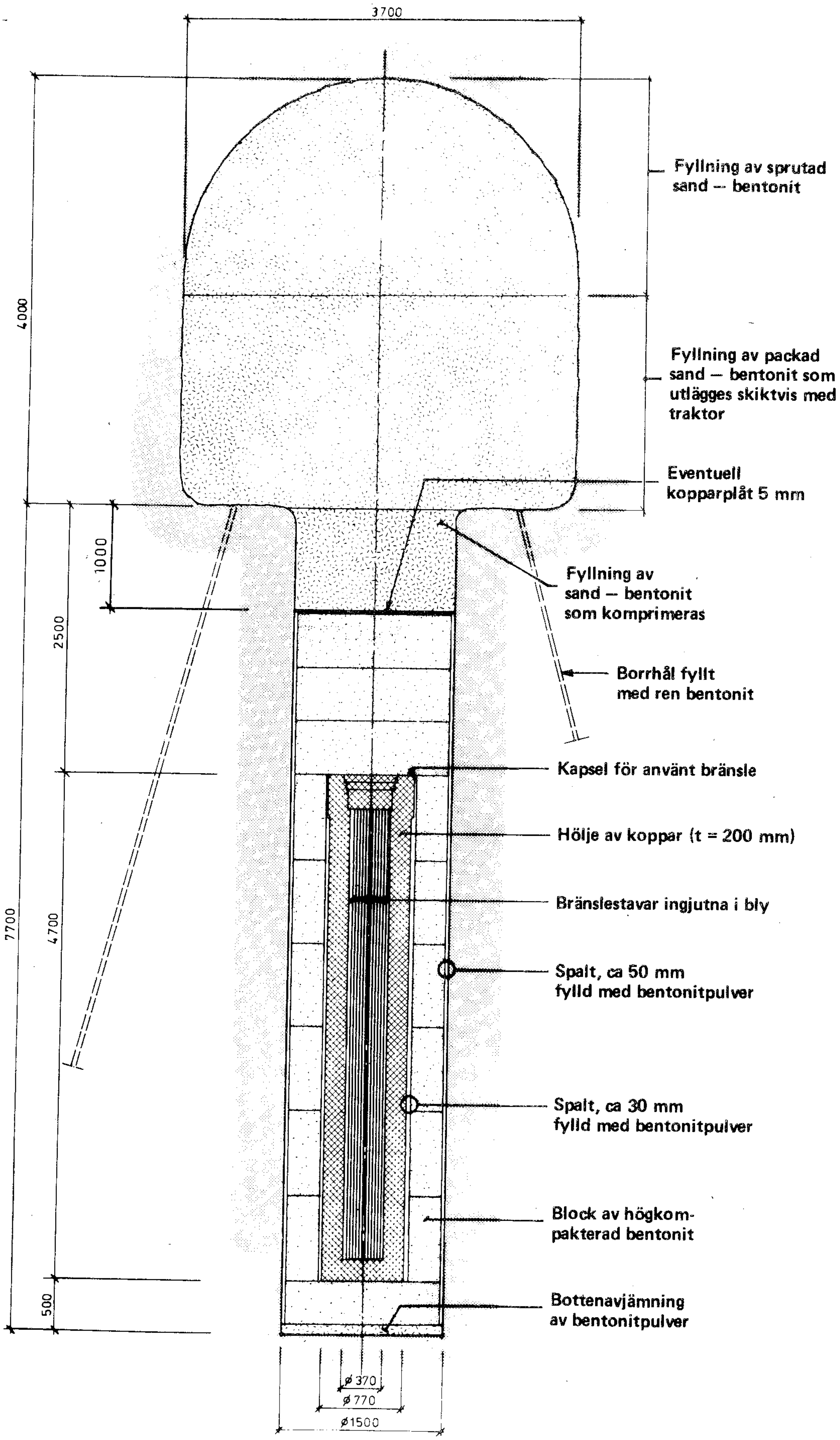

3700
4000
Fyllning av sprutad sand – bentonit
Fyllning av packad sand – bentonit som utlägges skiktvis med traktor
Eventuell kopparplåt 5 mm
1000
2500
Fyllning av sand – bentonit som komprimeras
Borrhål fyllt med ren bentonit
Kapsel för använt bränsle
Hölje av koppar (t = 200 mm)
Bränslestavar ingjutna i bly
7700
4700
Spalt, ca 50 mm fylld med bentonitpulver
Spalt, ca 30 mm fylld med bentonitpulver
Block av högkompakterad bentonit
500
Bottenavjämning av bentonitpulver
ø370
ø770
ø1500

FIG. 1

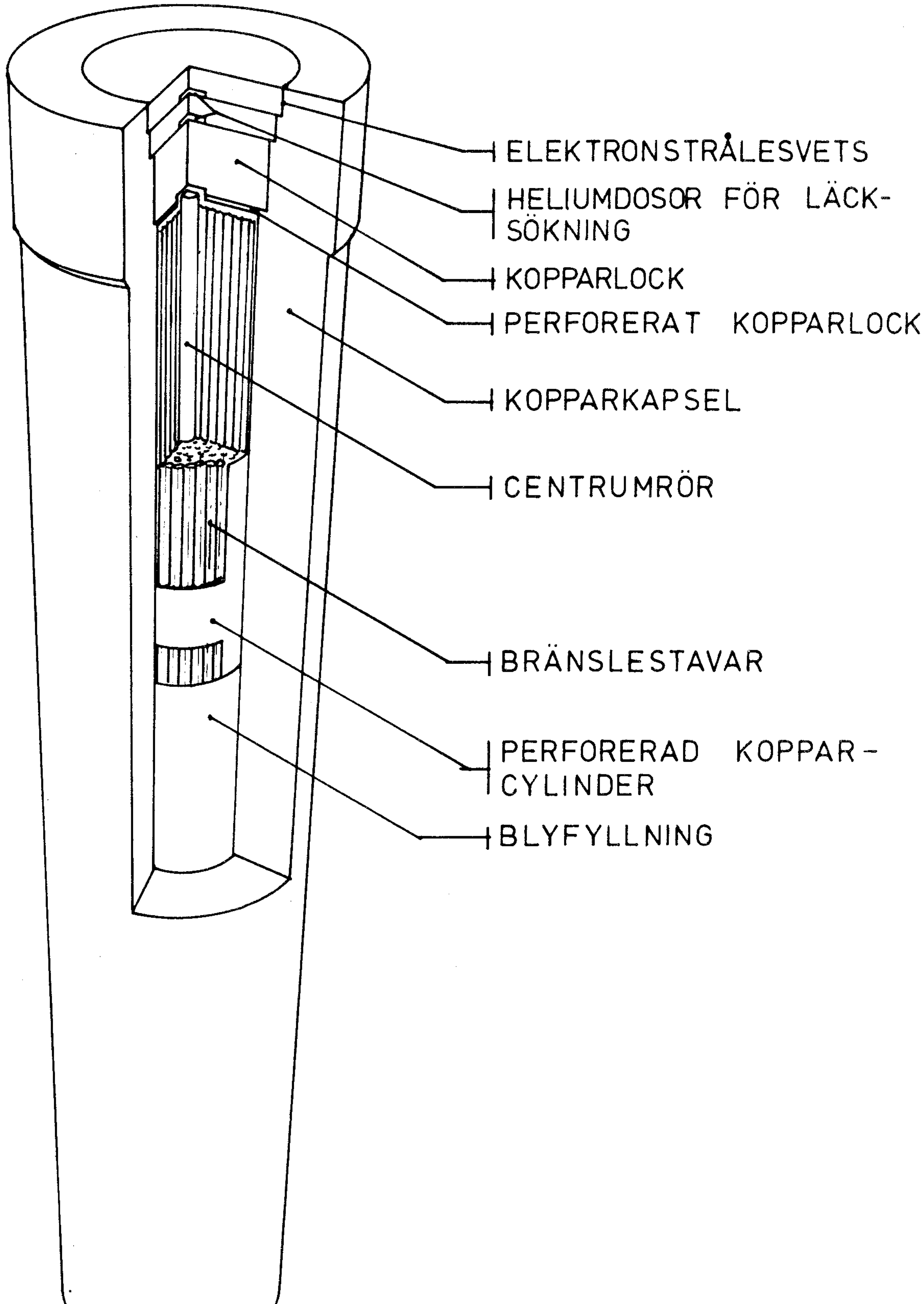

ELEKTRONSTRÅLESVETS
HELIUMDOSOR FÖR LÄCKSÖKNING
KOPPARLOCK
PERFORERAT KOPPARLOCK
KOPPARKAPSEL
CENTRUMRÖR
BRÄNSLESTAVAR
PERFORERAD KOPPARCYLINDER
BLYFYLLNING

FIG. 2

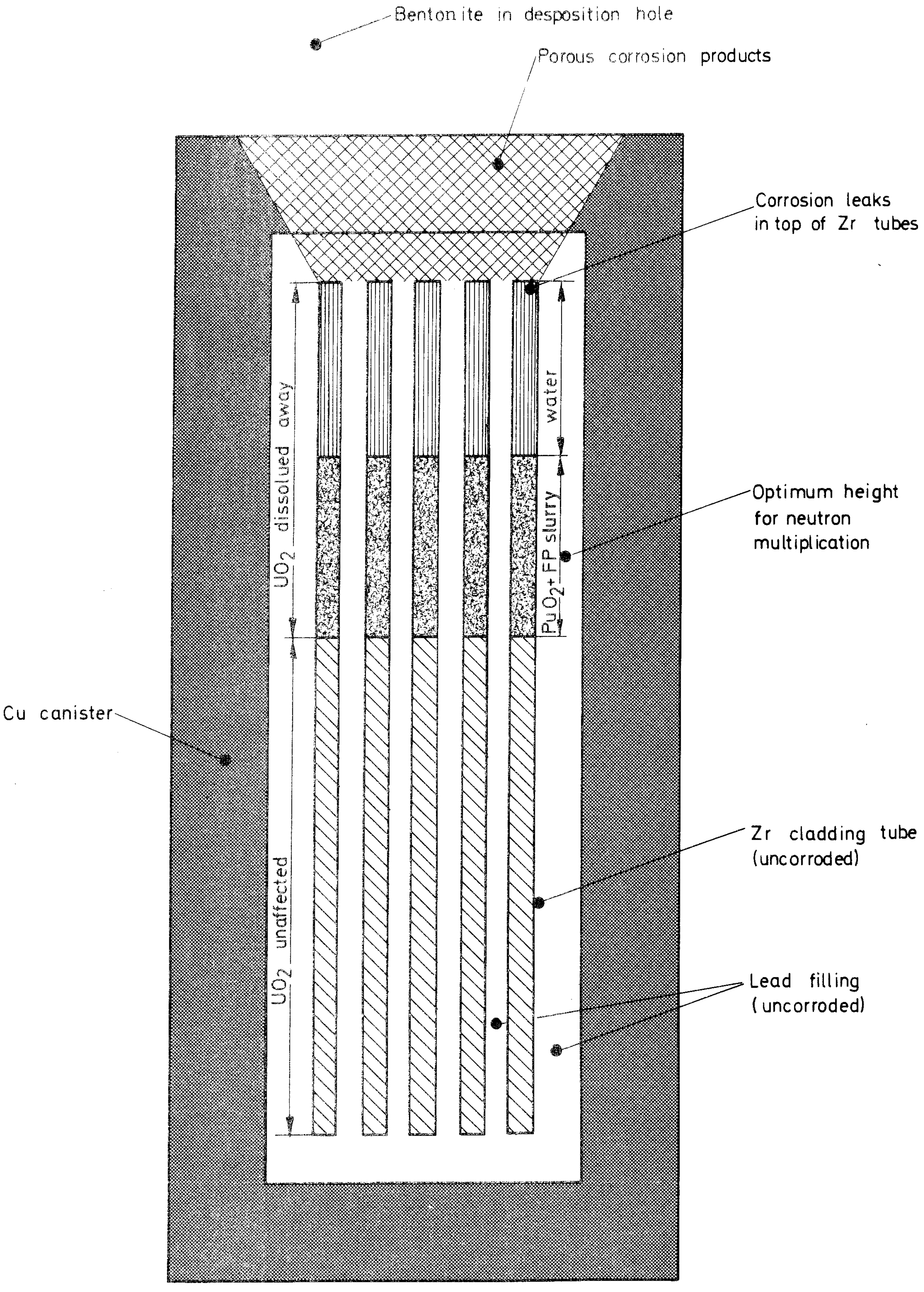

Bentonite in desposition hole
Porous corrosion products
Corrosion leaks in top of Zr tubes
UO_2 dissolved away
water
Optimum height for neutron multiplication
PuO_2+ FP slurry
Cu canister
Zr cladding tube (uncorroded)
UO_2 unaffected
Lead filling (uncorroded)

Fig. 3

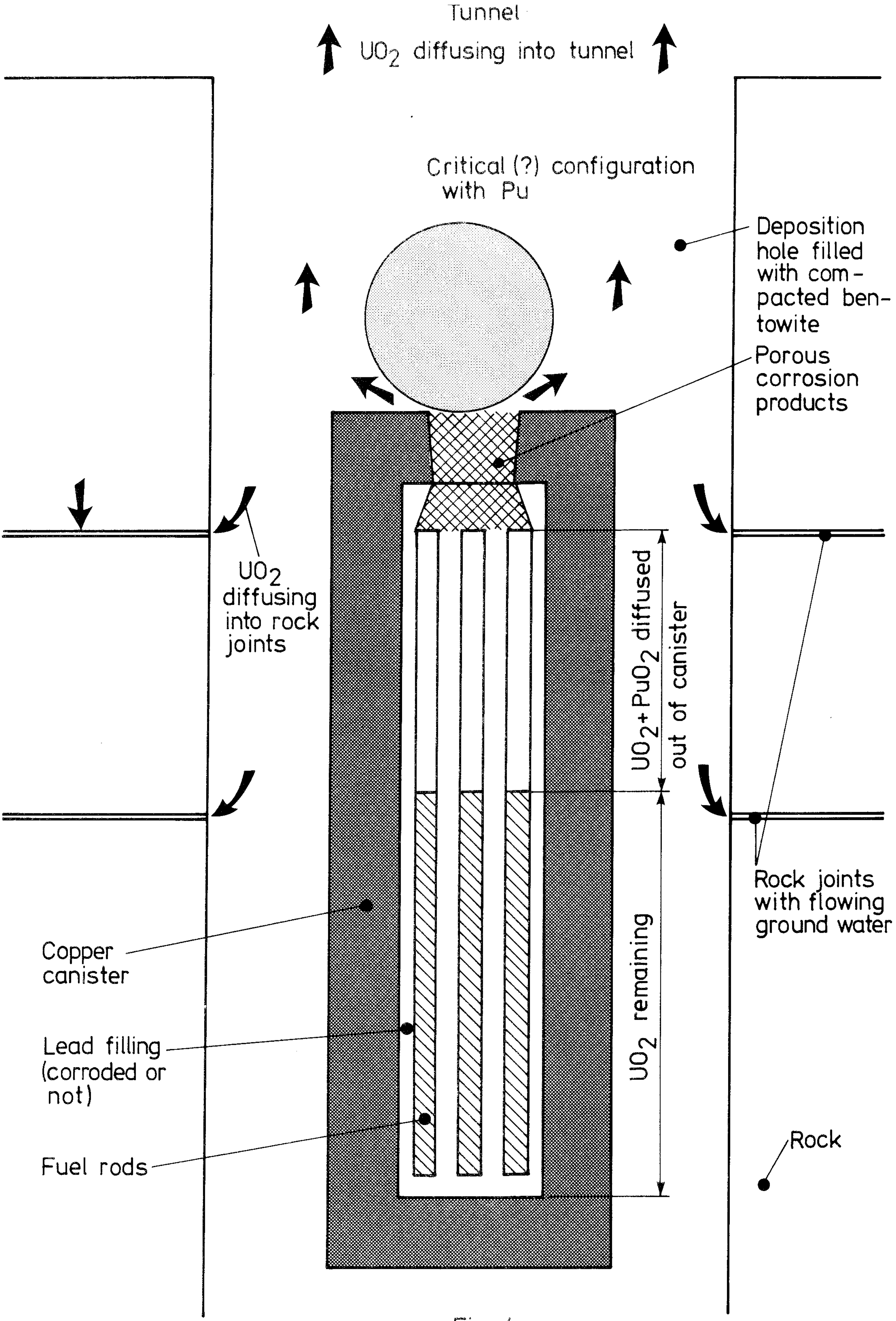

Tunnel
UO2 diffusing into tunnel
Critical (?) configuration with Pu
Deposition hole filled with compacted bentowite
Porous corrosion products
UO2 diffusing into rock joints
UO2+PuO2 diffused out of canister
UO2 remaining
Rock joints with flowing ground water
Copper canister
Lead filling (corroded or not)
Fuel rods
Rock

Fig. 4

Appendix 1 Processes occurring subsequent to canister penetration

"Canister penetration" is defined here as the formation of a hole through the 200 mm thick copper layer, so that contact is established between the water in the buffer material and the lead filling inside the canister. In order to occur within a time period when a major fraction of the Pu 239 remains, this would have to be a fabrication defect. The dimensions of the penetration cannot be defined here, but it must by necessity be a narrow pore or crack.

1 Copper corrosion

A very slow corrosion of copper (corresponding to a thickness reduction of the canister with 1-3 mm per 10^6 years) may occur continuously even before canister penetration. The initial rate may be somewhat higher due to presence of small quantities of atmospheric origin oxygen etc.

The initial result of penetration of the copper layer will be exposure of the lead filling. This will if anything provide anodic protection for the copper, perhaps resulting in the reduction of copper corrosion products at the canister surface.

Significant further corrosion of copper will occur only if and when oxidizing compounds (such as radiolytically produced O_2 or H_2O_2) start to diffuse out of the canister at a later stage. Until then the narrow passage constituting the initial penetration (weld crack etc) will remain unchanged.

Such further copper corrosion is possible only if the hydrogen arriving in at least stoichometric quantity with the oxidants remains chemically passive, since otherwise copper corrosion products would be reduced to metal. The latter appears likely but cannot be proved. The most conservative assumption is that all O_2 and H_2O_2 produces Cu_2O as a porous layer. As mentioned, this should be possible only after the lead is more or less completely oxidized, since otherwise the latter will provide anodic protection to the copper.

The canister will never "disappear" as a result of corrosion or dissolution reactions. It can only be converted to insoluble, perhaps porous, corrosion products (copper oxides and or sulphids) to an extent that is difficult to define. Any corrosion will increase its volume. Major changes of shape can occur only due to collapse caused by the swelling pressure of the bentonite after a free volume of water has formed inside. (Demolition due to tectonic movements is disregarded). Thus the volume in the deposition hole can be divided in three "compartments", i.e. the volume outside the canister, the canister itself (corroded and porous or not) and the volume inside the canister. Criticality could possibly occur due to concentration in either the first or the last mentioned "compartment".

2 Lead corrosion

As mentioned the lead will be anodic in relation to the copper, and the corrosion rate will most likely be limited by the indiffusion of anions while Cu corrosion products on the outer surface of the canister serve as electron acceptors. A precipitate, probably of lead sulphate, will form in the passage in the copper canister. In practice this precipitate will almost certainly block the process so that it soon comes to a complete stand still. However, without experimental evidence for this it must be conservatively assumed that this does not happen and that no effective blocking occurs. In this way a significant amount of lead could corrode in a relatively short time, and in the most unfavourable case water could establish contact with all the fuel rods in the canister via porous lead corrosion products, perhaps in a matter of decades or centuries.

During the time when the zircaloy in the fuel rod cladding tubes corrodes it provides anodic protection for the lead (unless it remains in the passive state). When the zircaloy is gone further corrosion due to radiolytically produced oxygen or H_2O_2 originating in the fuel zone may occur. As mentioned above the lead should be essentially completely oxidized before significant attack on the copper from this source is possible.

3 Zircaloy corrosion

Corrosion of the zircaloy cladding at low temperature is expected to be very slow. A corrosion rate of less than 1 μ in 100 years at 90°C is quoted 3). This would give a time to penetration of 50 000 - 80 000 years. However, such figures cannot be conservatively assumed. In view of the fact that we are dealing with an environment where the absence of pitting attack has not been proved it seems advisable to assumed a time for cladding penetration of perhaps hundreds rather than tens of thousands of years.

Subsequent to zircaloy cladding penetration the whole of the interior of the fuel rod becomes accessible to water. A slow but inexorable corrosion of the inside of the zircaloy cladding will start. While its rate may be low enough to require perhaps 100 000 years or more for completion, there is little quantitative basis for a precise estimate. The fact that the hydrogen will be largely confined to the corrosion zone may result in zirconium hydride formation, the break up of the protective oxide layer and fast attack. It seems prudent to assumed here that the time required for zircaloy corrosion to proceed to completion is small compared to the half life of Pu 239.

The corrosion of zircaloy will give rise to a stoichiometric amount of molecular hydrogen. The quantity involved for a single canister with BWR fuel is no less than about 186 Nm^3.

4 Radiolysis of water

Ionizing radiation will cause water radiolysis, involving formation of hydroxyl radicals and solvated electrons. These will react with each other producing hydrogen, oxygen, hydrogen peroxide and reaction products with other materials present. The outcome will depend primarily on the amount and kind of solutes present and the LET (Linear Energy Transfer) of the radiation.

Low LET radiation such as gamma radiation will promote the recombination of the radiolysis products into water, particularly in the presence of excess molecular hydrogen even in quite low concentration. This means that if the oxygen is consumed in an oxidation reaction with a solute the resulting excess of hydrogen will counteract further radiolysis, so that a quasi-equilibrium situation is reached 4). Further progress of the radiolytically originated oxidation reaction can therefore not proceed unless the hydrogen diffuses out of the reaction zone. (However, this is somewhat dependent on the details of the reaction kinetics).

In contrast to this, with high LET radiation, such as alpha radiation, recombination reactions are less favoured and net radiolysis will occur even in the presence of large concentrations of hydrogen 5). This means that the rates of radiolytically originated oxidation reactions will not be limited by diffusion constraints as for low LET radiations.

In the case of the waste canister this means that prior to contact between water and the fuel proper oxidation through radiolysis will give an insignificant contribution to corrosion rates etc. Following penetration of the zircaloy cladding the situation will become different, however, since the water in direct contact with the fuel in cracks and crevices in the latter will be exposed to alpha-radiation. The occurrence of oxidation reactions along with continuous evolution of free hydrogen gas can therefore not be ruled out. The rate of water radiolysis under various assumptions about the involvement of the oxygen and hydrogen peroxide in oxidation reactions is treated in ref (5).

5 Reaction of the fuel matrix subsequent to water ingress

Experience (from long term under water storage of damaged irradiated fuel) shows that this reaction is slow at least in relation to normal time scales involving human activities 3).

This is also confirmed by the results of leaching experiments where the rate of leaching from irradiated fuel fragments exposed in oxygenated water has been measured 6) 7). However, these results are hard to apply to the case under consideration, because of very different environmental conditions.

A prediction of the actual long term behaviour of the fuel following water ingress will be largely conjectural. Because of the confined conditions, it is reasonable to assume that oxygen and hydrogen peroxide formed by alpha radiolysis will be consumed by the uranium matrix.

Initially, the fluorite lattice of the latter may become hyperstoichiometric but eventually it will break down and reform into a new more oxygen rich lattice 8). In that connection fission products frozen into the original lattice will be freed and can start migrating out. Plutonium, substitutionally dissolved in the UO_2 will probably enter the lattice of any new UO_x-phase

The formation of higher uranium oxides is possible only if the excess hydrogen formed is chemically passive and leaves the reaction zone as gas. This may not be the case, since the UO_2 contains e.g. small quantities of platinum metal fission products such as palladium that may serve as reduction catalysts. If this occurs, there may be no net oxidation and dissolution of uranium, although the original UO_2 matrix may be broken up. It should be observed that the presence of hydrogen from zircaloy corrosion may further counteract any oxidation reaction, although the time interval when this occurs cannot be fixed.

A theoretical calculation of alpha radiolysis in the case that oxygen and hydrogen peroxide formed are promptly consumed by the UO_2 matrix5), and assuming that the volume of water filled alpha-irradiated fine pores in the fuel is 5 % of the fuel volume (a very high figure) gives a time to oxidation of the matrix to a composition corresponding to UO_3, or about 3 000 years.

This value is almost certainly on the low side, probably by orders of magnitude, but should conservatively be assumed here.

Whether oxidized or not the uranium oxide and its content of actinides is practically insoluble in water and will not be transported away by diffusion as such. Only to the extent that they can form soluble complexes with incoming anions, primarily bicarbonate ion, can significant diffusional transport occur.

Hexavalent uranium is known to form such soluble complexes. On the basis of maximum assumed bicarbonate content in the water in the buffer material outside the canister and the dimensions and properties of the diffusion path the maximum amount removed from the fuel zone can be estimated.

With tetravalent uranium solubilities and transport rates are 3-4 orders of magnitude lower and diffusional transport will be negligible in the time span of interest for plutonium criticality.

Plutonium is likely to remain in the tetravalent form when and if hexavalent uranium is formed, thereby largely remaining behind as the latter diffuses away. This creates the potential for separation that is a prerequisite for criticality conditions. In the unlikely case that plutonium is oxidized to the hexavalent stage and forms soluble complexes, it will diffuse away along with the associated uranium of the oxide matrix it was contained in. Separation of the two is then possible in principle by preferential reduction of the plutonium along the diffusion path.

Leaching of uranium in the form of hexavalent carbonate complexes (primarily $UO_2 \cdot (CO_3)_2^{2-}$ and e.g. subsequent precipitation by reduction in the buffer material zone may in principle create a free water volume inside the canister. x)

x) In reality a number of soluble uranyl examples will exist in addition to this, such as $UO_2 \cdot (CO_3)(OH)^-$, UO_2CO_3 and $UO_2(CO_3)_3^{4-}$, but for order of magnitude considerations it is sufficient to consider the dicarbonate complex.

At the same time the latter is exposed to outside pressure i.e. the swelling pressure of bentonite. The canister may or may not resist this pressure, depending on the extent of corrosion subsequent to penetration due to radiolytic oxygen production (which is likely to be unimportant but cannot be ruled out). In case of collaps of the canister the inside free water volume created by uranium extraction will of course more or less disappear and the corresponding outside volume created will be filled with bentonite gel due to the swelling of the compacted bentonite surrounding the canister.

The experiences from the Oklo phenomenon is that irradiated UO_2 (uraninite) has been stable for geologic periods of time in contact with ground water, thus strongly contradicting the above scenario involving UO_2 lattice break down and dissolution. However, the (partly unknown) difference in environmental conditions makes it impossible to draw any definite conclusions from these observations.

6 Conditions in the buffer material

The conditions in the buffer material before canister penetration have been described in section 2.

Canister penetration will initially have no consequences until net hydrogen gas production from zircaloy corrosion, plus a possible gas production from alpha-radiolysis of water in the fuel zone starts.

The diffusion coefficient of hydrogen in compacted bentonite has been determined experimentally 9). Using the value thus obtained it can be shown that it would take at least 500 000 years under the most favorable conditions for the hydrogen created to diffuse away from the deposition hole into the above laying-tunnel or into the rock joints.

Thus, unless extremely optimistic assumptions are made concerning the rate of corrosion of the zircaloy, formation of a gas bubble in the compacted bentonite cannot be avoided. The mechanical effects of this are discussed in ref (10).

The bubble may or may not disappear by making direct contact with water filled joints in the host rock outside the bentonite filled zone. This could only occur through some type of mechanical movement in the rock. A very minor movement may be sufficient, and the disappearance of the gas could be quite rapid.

Following release of the gas pressure in this way, the bentonite will expand back into the resulting void by swelling. In the end the original situation will be largely restored, with somewhat higher water ratio in the bentonite due to hysteresis effects. Free water volumes will never exist in the deposition hole outside the canister.

7 Transport of uranium in and from the buffer material

As mentioned in section 5 uranium in the fuel mass may be dissolved as uranyl carbonate complex following oxidation to the hexavalent form. Migration of major amounts of uranium from the canister, sufficient to set enough plutonium free to achieve a criticality, requires that the carbonate complexes are removed from the immediate vicinity of the canister either by diffusion or by precipitation in the soluble tetravalent form. The quantities that can remain in solution and adsorped in the hexavalent stage in the bentonite in the deposition hole are much too small in this respect.

Material diffused from the canister can be removed and disappear either through the cracks in the host rock or into the tunnel above the deposition hole. In the latter relatively large amounts of ferrous iron are present for desoxidation purposes, and so the potential exists for precipitation of tetravalent uranium. The buffer material in the deposition hole contains only small amounts of ferrous iron 11) (0.12 %) and reaction of the total amount corresponds to a uranium reduction from hexa- to tetravalent form (and precipitation) of 65 kg U.

The material that can be held up in the bentonite in hexavalent form can be estimated on the basis of absorption equilibrium established in other investigations.

The quantities that can disappear through cracks in the host rock can be calculated using data and calculation methods in provided by Neretnieks 9) 12).

An upper limit of rate of diffusion from the vicinity of the canister into the tunnel, assuming the latter to be an infinite sink for uranium, can be made straightforwardly by assuming a linear concentration gradient of uranyl carbonate complex concentration from saturation (1070 g/m^3) at the upper end of the canister to zero at the floor of the tunnel.

Transport processes may be influenced by the presence of gas volumes within the buffer material. These will block diffusion within the buffer material as long as they persist. The extra pressure caused by the gas may open up the joints in the rock somewhat, and the bentonite may have a higher water content than originally if the bubble escapes and is replaced by water expanded bentonite. Both these effects have been taken care of in the selection of diffusion coefficient and joint pattern in calculating the figures given below.

The diffusion coefficient for the large uranyl carbonate ion in the bentonite has been assumed to be as high as $8 \cdot 10^{-11} m^2/s$ and the crack pattern is assumed to be one 0,2 mm crack every 0.4 m (corresponds to a permeability some 10^4-10^5 times that initially in the repository region.)

Calculated values for amount of uranium removed from the canister, are shown in the following table 1. It should be observed that the values given are intended to be conservative rather than realistic.

Table 1

Maximum amounts of uranium that can be transported from a leaking canister as uranyl carbonate complex. Concentration of the latter assumed to be 1070 g/m^3 at the canister surface.

Time after disposal years	Hold up in solution in depos. 1) hole kg	Amount reduced by Fe^{2+} present in depos. hole 2) kg	Disappeared through joints in host rock 3) kg	Disappeared into tunnel kg	Total kg
0	100	65	0	0	165
10^4	100	65	31	20	216
$3 \cdot 10^4$	100	65	93	60	318
$6 \cdot 10^4$	100	65	186	120	471
10^5	100	65	310	200	675

1) Assuming 30 vol % water

2) These contributions are assumed to be instantaneous although in reality that is not the case.

3) These amounts would probably clog the joints.

4) Assuming infinite capacity for reduction of hexavalent uranium to tetravalent.

Appendix 2 Criticality with plutonium

1 Reactivity of plutonium inside a canister

In the spent fuel repository significant amounts of plutonium are buried in the canisters. If there is a leak in a canister uranium plus fission products could diffuse away leaving the plutonium, lead and zircaloy behind. This diffusion-process must occur in sufficiently short time, so that the plutonium has not yet decayed to uranium.

In the calculations reported here we have assumed that all uranium and all fission products are removed from the canister instantaneously. The lead filling and the zircaloy cladding tubes remain unchanged. The inside of the latter becomes filled with water instead of UO_2-pellets. The plutonium left behind mixes with this water.

Normally one would expect that the plutonium would sink to the bottom of the canister in a form of an oxide-slurry, but this is not assumed here. Instead we have assumed that the plutonium mixes homogenously with the water giving well moderated configurations.

We have calculated the reactivity of these configurations as a function of time and plutonium concentration using the computer program MICO, which calculates critical parameters for homogenous media. Table 1 gives the result of the calculations.

Both the amount of plutonium and the isotopic composition change with time. These effects initially roughly compensate each other, since the decay of Pu 239 is compensated by the faster decay of the neutron absorber Pu 240. The results are given for a number of different times after disposal.

Many configurations in table 1 have a keff less than 1.0, which means that criticality is not possible for these configurations. For those configurations for which keff is larger than 1.0 for a canister of infinite height, the actual critical height is calculated. This critical height is compared with the "available height" i.e. with the height that can be achieved with the plutonium available at the instant of time. As can be seen the available height is always less than the critical height. That means criticality is not possible in the canister for any configuration at any time. Furthermore we have to realize that not all fission products are soluble. The contribution of the unsoluble fission products to the reactivity is about -0.05 to -0.10, reducing the multiplicating factor even further.

Table 1

Critical parameters[x)] of Pu configurations inside a canister

Time (a)	Pu density in water (g/cm^3)	keff (infinite height) of the canister	critical height (cm)	available height (cm)
0	0.05	0.883	-	-
	0.10	0.939	-	-
	1.00	0.871	-	-
	5.00	1.167	29.7	0.3
	11.46	1.550	9.8	0.15
6000	0.05	0.936	-	-
	0.10	0.991	-	-
9000	0.05	0.957	-	-
	0.10	1.014	164.4	17.3
24000	0.05	1.028	110.8	35.5
	0.10	1.093	50.4	17.7
30000	0.05	1.035	96.5	34.2
	0.10	1.096	49.1	17.1
40000	0.05	1.047	81.2	30.9
	0.10	1.109	44.7	15.4
50000	0.05	1.049	78.9	27.2
	0.10	1.110	44.4	13.6

x) Critical parameters are calculated with a reflector saving of 11.0 cm.

2 Reactivity of plutonium in the buffer material outside the canister

In the spent fuel repository significant amounts of plutonium are buried in the canisters. If there is a leak in a canister plutonium can diffuse away and might accumulate outside the canister in the surrounding bentonite. Here we assumed that not enough time has passed, so that the plutonium has not yet decayed to uranium. This also means that plutonium from several defect canisters cannot unite. Such a process would require many plutonium half-lives.

The bentonite has a water content of 29.6 %. In this water PuO_2 can accumulate. It is assumed that no other constituents like uranium or fission products are present.

The amount of fissile plutonium in one canister is about 9 kg at the time of the disposal and is decreasing with time. But, because Pu 240 has a shorter halflife than Pu 239, the minimum critical mass is decreasing with time until Pu 240 has decayed completely.

We have calculated the critical mass as a function of time and plutonium concentration using the computer program MICO, which calculates critical parameters for homogenous media. In these calculations it was assumed that all plutonium from one canister is collected outside the canister in the bentonite in a spherical configuration. Table 2 gives optimum plutonium concentration and minimum critical radius and critical mass for each timestep. As table shows the minimum critical mass is obtained for well moderated critical configurations. That means it is not possible to reach criticality without water, i.e. in a "fast" system. To go critical without water would in bentonite require more than 100 kg of plutonium, i.e. more than 10 times as much as there is plutonium in one canister. That means, even if a critical configuration has been achieved the power level is limited by the requirement that the heat must be conducted away otherwise the water would boil away and leave the configuration subcritical. An approximate calculation of the power level in the critical sphere using the arguments given in appendix 3 gives maximally about 5 kW.

If the sphere is in thermal connection with the canister heat will be conducted away from the surface of the cylinder. In this case the power level could be larger by a factor of roughly 4, i.e. 20 kW.

The comparison of the critical mass and the canister inventory shows that any criticality will have to take place within the first 50 000 years.

If the bentonite in the deposition hole is totally or partly removed, the critical mass will obviously decrease, as shown in table 3, which refers to a time after disposal of 30 000 years.

Table 2 : Pu critical spherical configurations in bentonite.

Time (a)	Pu density in water (g/cm^3)	Critical[x] radius (cm)	Critical mass (kg Pu-239)	Pu-239 mass in one canister (kg)
0	0.05	48.9	5.3	8.9
10 000	0.05	39.2	3.2	6.7
30 000	0.04	36.2	2.3	3.8
60 000	0.04	35.5	2.2	1.6
100 000	0.04	38.4	2.8	0.5

x) The critical radius was calculated with a reflector saving of 11.8 cm.

Table 3

Minimum critical mass as function of percentage of bentonite lost from the deposition hole at 30 000 years after disposal.

Percentage loss	Minimum critical mass, kg
0	2.3
25	1.3
50	0.8
75	0.6
100	0.5

As previously mentioned, thermal limitations place an upper limit of 20 kW on the "reactor power" corresponding to a temperature of 265°C at the canister surface.

It is of some interest to know the upper limit of the total energy generation (in other words the amount of radiotoxic nuclides) that could be produced by such a "reactor". Clearly an absolute upper limit must be that corresponding to all plutonium initially present minus the minimum critical mass.

The minimum possible mass is obtained, when the amount of bentonite is just able to keep the tunnel filling from falling down into the deposition hole (corresponding to 90 % bentonite lost). (An invasion of quartz sand would strongly increase the critical mass). Interpolating from table 3 this gives ∿0.5 kg as the critical mass.

Using the arguments presented in appendix 1 section 7, and doubling the figures for uranium disappeared into the tunnel by reason of the increase in diffusivity resulting from the "thinning" of the bentonite, a critical configuration could form at 20 000 years after disposal of the spent fuel. At this time 3.9 kg fissile plutonium originating in the canister remains. An absolute upper limit of the energy generation is obtained by assuming that all of it minus the critical mass is fissioned. This gives an energy release of ∿3 000 MWd, i.e. 7 % of the energy released by the spent fuel placed in the canister.

Calculation of the reflector saving

The critical parameters in appendix 2 and 3 are mostly calculated using fundamental mode theory, i.e. zero-dimensional calculations. In zero-dimensional calculations the critical buckling is calculated. The critical radius of a sphere or an infinite cylinder can be calculated from the critical buckling B_c^2.

$$R = \frac{c}{\sqrt{B_c^2}} - \delta$$

where c is a geometry dependent (known) constant and δ is the reflector saving. If for one special case both zero- and one-dimensional calculations are done, the reflector saving can be calculated for this case. This reflector saving was then used for all other cases of the same problem. As can be seen from tables 1 and 2 in app. 2 and table 6 in app. 3 the reflector saving varies very slightly between different problems.

Therefore the use of the same reflector saving in different cases of the same problem seems justified. (The reflector saving was always calculated for the most important case of a problem. So for the most important case the reflector saving is correct).

Appendix 3 Criticality with uranium 235

In the spent fuel repository large amounts of uranium and plutonium are buried in the canisters. If there are leaks in the canisters uranium and plutonium can diffuse away and might accumulate in the tunnels. Here we assume that sufficient time has passed, so that the Pu 239 has decayed to U 235. The enrichment in U 235 of the uranium is then 1.66 % according to the assumptions made about the type of spent fuel in the canisters. (PWR-fuel irradiated to 33 000 MW/t). Criticality could take place either in the tunnels or the deposition holes.

1 Criticality in the tunnels

When the uranium with this enrichment is accumulated in the sand of the tunnel and water is present criticality is theoretically possible. The calculations have been done with the computer program MICO, which calculates critical parameters for homogenous mixtures. If the uranium is assembled in a spherical volume the calculated minimum critical mass is 4.4 t, when there is no power generation. When power is generated more uranium is needed to keep the configuration critical. The spherical configuration has a radius of about 0.9 m and a volume of 3.5 m^3. If the volume is not spherical the minimum critical mass will be larger.

In the following we will assume that uranium has accumulated somewhere in the tunnel to form a critical configuration and will investigate its consequences.

If criticality has been reached power generation will start and the temperature of the configuration will rise and reduce the water density. With decreasing water density reactivity will decrease (see tab. 4). (Negative moderator density coefficient). That means the power level will be limited. If during the passage of time more and more uranium is accumulated the power level must go up, so that the reduced water density keeps the configuration just critical. (Because of the slow diffusion of uranium in the sand and because of the negative moderator density coefficient, the critical configuration will always be in a state of quasi-equilibrium.)

The total power of the critical configuration will be given by the amount of cold water flowing past the configuration and by the heat conduction away from the configuration to the outside. (Heat must in some way be removed from the critical configuration, otherwise the water would boil away and the dry configuration would be subcritical stopping all further power generation). As the flow of water through the sand is very slow, the heat conduction will mainly determine the power level.

In the tunnel, the largest critical configuration with the largest possible total power would be a cylindrical configuration with the radius equal to the radius of the tunnel and the length of the cylinder given by the total amount of uranium in the tunnel. I.e. the uranium from all canisters is assumed to be collected at one place with the uranium density so low that criticality is just possible. (We make here the extremely unlikely assumption that all uranium from all canisters in the tunnel is collected at one place and that in addition the uranium density is chosen to give the largest possible critical volume).

To calculate the power P_{tot} generated in this configuration we start from the equation of heat conduction.

$$P_{tot} = \kappa \int \text{grad } T \, df \qquad (1)$$

where the integral is taken over the surface of the critical configuration.

An analytical calculation of eq. (1) is only possible when making simplifications. The simplifications we will make, allow us to determine an upper theoretical limit to the total power generated in the tunnel.

A sphere has a larger temperature gradient across its surface than a cylinder other things being equal. We therefore replace the temperature gradient at the surface of the finite cylinder in eq. (1) by the temperature gradient at the surface of a sphere with the radius equal to the radius of the tunnel.

The temperature distribution outside a sphere in the surrounding rock is given by: (see "Carslaw and Jaeger, Conduction of Heat in Solids. Oxford Press 1969").

$$T\,(r,t) = T(\infty) + (T(r_0) - T(\infty))\,\frac{r_0}{r}\left[1 - \text{erf}\,\left(\frac{r - r_0}{2\sqrt{\frac{\kappa}{\rho c}\,t}}\right)\right] \qquad (2)$$

where:

$T(\infty)$ is the temperature far away from the spherical heat source.

r_0 is the radius of the spherical heat source.

erf is the error function

κ, ρ, c are heat conductivity, density and heat capacity of the surrounding rock.

$T(r_0)$ is the (timeconstant) temperature at the boundary of the spherical heat source, i.e. the boiling temperature of water at the appropriate pressure.

As the gradient of the temperature at the surface of a sphere by reasons of spherical symmetry always is directed in the radial direction, we get for grad T at the surface of the sphere $r = r_0$:

$$\left.\frac{\partial T}{\partial r}\right|_{r=r_0} = -(T(r_0) - T(\infty))\left[\frac{1}{r_0} + \frac{1}{\sqrt{\pi\,\frac{\kappa}{\rho c}\,t}}\right] \qquad (3)$$

The second term in the square bracket tends to go to zero for large times t. It becomes negligible compared to the first term for times satisfiing:

$$t >> \frac{r_0^2 \rho c}{\P \ \kappa} \qquad (4)$$

Taking the actual data for the tunnel and the surrounding rock this means times larger than a few years.

Introducing eq. (3) into eq. (1) we get for P_{tot} the upper limit

$$P_{tot} = \kappa(T(r_0) - T(\infty)) \frac{1}{r_0} \cdot F \qquad (5)$$

where F is the surface of the critical configuration. As we see P_{tot} is constant in time because we disregarded the second term in the square bracket of eq. (3), which was justified for times greater than a few years. (Because the diffusion of uranium will be extremely slow, we will in fact always be in a state of quasi-equilibrium, that means all time variations will be extremely slow).

The amount of fission products in the critical configuration depends on the total power and the half-lives of the fission products. In the steady state we have the equation:

$$N(\text{fissproduct}) = \frac{1}{\lambda} \ y \ \frac{P_{tot}}{E_f V} \qquad (6)$$

where:

N is the average number density of the fission product
λ is the decay constant
y is the fission yield
E_f is the energy per fission
V is the volume of the critical configuration.

Numerical calculations

We shall use the data given in tab 1 and 3 to calculate actual results for our problems.

With the total amount of uranium 227 ton U in the tunnel and a minimum critical density of 0.78 ton U/m^3 the maximum critical volume is 291 m^3. We assume that the critical configuration consists of a finite cylinder with the radius of the tunnel 1.85 m. The length of the cylinder then is 27 m and its surface 335 m^2. An upper limit for the total power generated is then given by eq. (5)

$$P_{tot} = 3.0(265-20) \frac{1}{1.85} 335 = 1.3 \cdot 10^5 \text{ W} = 130 \text{ kW}$$

The power per unit of length of the cylinder is

$$p = \frac{130}{27} = 5 \text{ kW/m}$$

It should be pointed out once again that the total power given is an upper limit. If we assume that the finite cylinder has the same temperature gradient as an infinite cylinder except for the ends of the cylinder the total power would be lower by a factor of about 3, i.e. 40 kW.

To give an example of the fission product number density we use eq. (6) to calculate the average number density of Sr 90 in the critical configuration.

$$N(\text{Sr }90) = \frac{1}{7.5 \cdot 10^{-10}} \; 0.0591 \; \frac{1.3 \cdot 10^{5}}{3.24 \cdot 10^{-11} \cdot 291 \cdot 10^{6}} =$$

$= 1.1 \cdot 10^{19}$ atoms of Sr 90/cm^3.

This is equal to 250 Ci of Sr 90 per meter of generating length, a figure that should be compared to about 12 00 Ci/m in the canisters at the time of disposal.

Tab. 1 Data for the tunnel and the surrounding rock

Radius of the tunnel r_0	1.85 m
Heat conductivity of the rock κ	3.0 W/m^{o}C
Density of the rock ρ	2700 kg/m^3
Heat capacity of the rock c	850 J/kg^{o}C
Pressure at 500 m depth	5 MPa
Boiling temperature at 5 MPa $T(r_0)$	265 ^{o}C
Temperature "far away" $T(\infty)$	20 ^{o}C

Tab 2. Composition of the sand in the tunnel (85 % sand + 15 % bentonit)

SiO_2	1.55 g/cm^3
Al_2O_3	0.04 "
Fe_2O_3	0.01 "
Porosity x)	42 %

x) It is assumed that the porosity (void volume) is filled with a mixture of water and UO_2. The ratio UO_2 to water is varied.

Tab 3. Criticality data for uranium

U-235 enrichment	1.66 %
Minimum critical mass (zero power)x)	4.4 tU
Error limits	3.0<M<7.2 tU
Minimum critical uranium density (k_∞=1.0)	0.78 g/cm^3
Mass uranium per canister	1.42 t
No of canisters in the tunnel	160
Total mass of uranium in the tunnel	227 t

x) At max. possible power the critical mass would be more than 4 times larger, i e about 18 tU.

Tab. 4. Influence of the water density on the reactivity (ρ(U)=1.26 gU/cm^3)

$\rho(H_2O)$	k_∞	k_{eff} x)
1.0	1.079	1.000
0.75	1.066	0.963
0.50	1.022	0.883
0.25	0.898	0.705

x) constant buckling $B^2 = 10.0 \cdot 10^{-4}$ cm^{-2}

Tab. 5. Data for the calculation of Sr-90 number density

Half-live of Sr-90	29 a
Decay constant λ of Sr-90	$7.5 \cdot 10^{-10} s^{-1}$
Yield of Sr-90 per fission of U-235	0.0591
Energy per fission of U-235 E_f	$3.24 \cdot 10^{-11}$ Ws

2 Criticality in the deposition holes

Using the calculation methods described above, the critical mass and radius for 1.66 % enriched uranium in a mixture of bentonite and water with varying water ratios has been calculated. The results are shown in table 6. By interpolation it is found that criticality is possible with the uranium inventory available in one canister with a water/bentonite ratio of 62/38.

This corresponds to a loss of 46 % of the original bentonite available.

An upper boundary of the amount of fission products and other radiotoxic nuclides that could be produced by such a criticality is obtained by making a burn up calculation of a reactor with its uranium inventory dispursed in a mixture of water to bentonite of 90/10. (Lower bentonite densities would not withstand the pressure from the material in the tunnel above and would collapse). It is found that this corresponds to an energy generation of 20 000 MWD. This means that the amount of radiotoxic nuclides generated will be about 43 % of the original inventory in the canister.

Table 6

Calculation of the minimum water to bentonite ratio that enables criticality with the uranium (1.4 t) of only one canister.

Water/bentonite ratio	Uranium density in the water (gU/cm^3) Critical mass (tU)		
50/50	2.0	2.5	3.0
	7.47	-	2.69
60/40	2.0	2.5	3.0
	2.48	1.55	1.50
70/30	2.0	2.5	3.0
	1.33	0.97	0.98

Interpolated minimum water to bentonite ratio is 62/38 i e 46% loss of the original bentonite.

Normal water to bentonite ratio is 29.6/70.4

The calculations are done with a reflector saving of 10.2 cm

Appendix 4 Influence of a local criticality on the corrosion of surrounding canisters in the repository

1 General

As mentioned in appendix 1 radiolysis resulting from contact between water and the irradiated fuel may result in the production of oxidizing species. The extent to which this takes place cannot be estimated with certainty at the present time, however. Transport of these oxidizing species to nearby canisters could in principle increase their rate of corrosion and shorten their life (time to penetration).

2 Oxidizing species

The following species have to be considered as possible oxidants of copper in case they are transported to neighbouring canisters.

a) Direct radiolysis products of water:
free oxygen and hydrogen peroxide.

b) Nuclides from the irradiated fuel in an oxidized stage.

c) Copper compounds from the failed canister in the divalent stage.

Regarding the importance and amounts of these products the following can be said:

- It must be assumed that free oxygen can occur until the reverse has been proved, although it appears unlikely. The upper limit of the concentration of dissolved oxygen will be reached if actual gas evolution takes place due to water radiolysis. This gas must then contain an at least stoichiometric amount of hydrogen, since the latter is assumed not to be used up in any side reactions, whereas this will always be the case with oxygen. Hence the upper limit for the partial pressure of oxygen is one third of the hydrostatic pressure in the repository, i.e. 1.7 MPa, and the maximum content of dissolved oxygen is that saturated at the above pressure and the prevailing temperature (∿500 ppm at 50°C).

- Hydrogen peroxide can be disregarded. It may originally be produced by radiolysis in fairly large amounts, but since in any passage out of the copper it must experience a long residence time in contact with copper oxides and other copper compounds it can safely be assumed that it will be catalytically decomposed to free oxygen and water.

- The principal (>95 %) dissolved component from the irradiated fuel is uranium, that appears as uranyl carbonate complexes. An investigation of the thermodynamics of its oxidation of copper is under way, and preliminary results show that it is not feasible. However, in the absence of final results it is assumed here that it acts as an oxidant. The upper limit of concentration is considered to be 1070 g/m^3 (12).

- Some fission products and actinides other than uranium can also occur in an oxidized form thermodynamically capable of corroding copper, e.g. plutonium (VI), neptunium (V), ruthenium, palladium and silver compounds etc. However, the quantities involved are too small for this to be of any importance.
- Divalent copper compounds could potentially form in very large quantities. However, at pH 8.5-9 and with carbonate present their solubility will be sufficiently small to eliminate them from consideration.

On the basis of these considerations free oxygen will be the dominating oxidating species by far.

3 Transport of oxidizing species

The mechanism of diffusional transport in the clay barrier and mass transfer from the latter to the flowing ground water in the rock joints has been described in ref (12).
Figure 1 adapted from figure 5 in reference (12) depicts the laminar ground water flow in a planar fissure perpendicular to the deposition hole and the build up of a concentration field of dissolved substance when the water flows past the hole. If as in the figure, another hole is directly downstream the process will be reversed there. An upper limit of the transport of oxidants into the downstream hole is obtained if it is assumed that their concentration in the ground water impinging on it is equal to the concentration inside the upstream hole containing the critical configuration.

As long as parts of the copper canister remain in the metallic state oxygen from the critical region will be used up by diffusion to those parts. However, if the fissure being considered for oxidant transport is opposite the critical configuration (e.g. as shown in figure 4 in the main report) this is of no consequence.

4 Numerical results

The calculations are made using a typical rock permeability of 10^{-9} m/s, and a hydraulic gradient of 0.5 %. This gives a bulk flow velocity of water through the rock of $2 \cdot 10^{-4}$ m/year. The permeability around the hole is assumed to be increased by a factor of 10^3 due to cracking resulting from excavation work etc, which corresponds to one 0.1 mm joint (fissure) every meter.

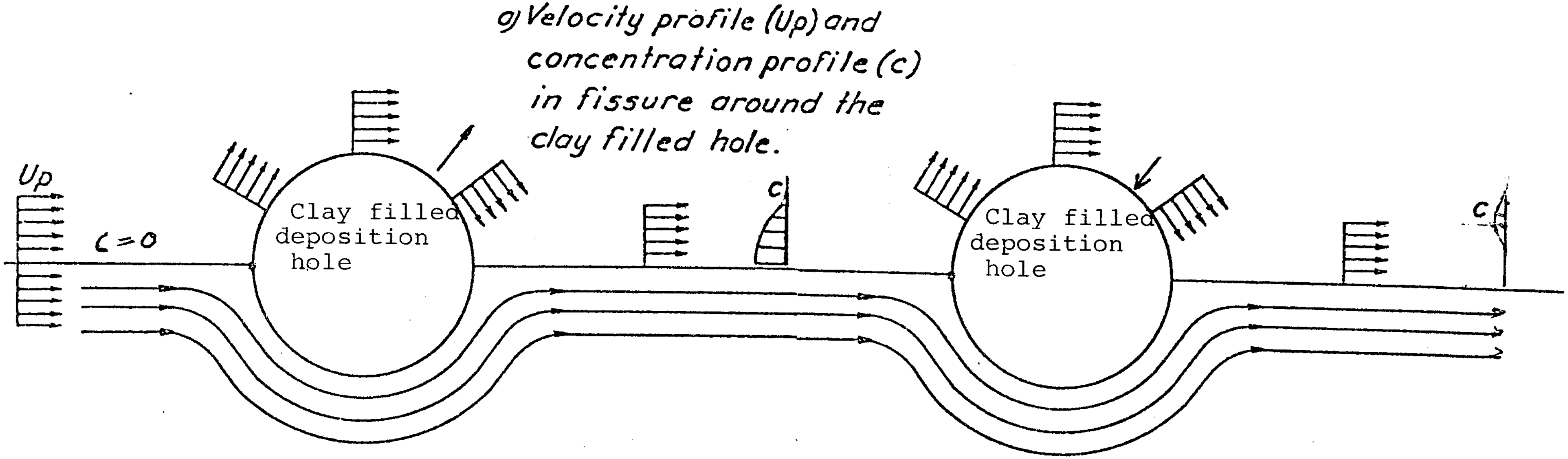
With the mentioned oxygen and uranyl carbonate concentrations this gives a time to average corrosion of 8 mm (4 %) of the thickness of the canister in 170 000 years. This is the time to penetration corresponding to the maximum pitting factor mentioned in ref. (2). It is seen that this time is $\sim 10^3$ times the time that the reactor can be "on power".

5 Conclusion

Even at maximum theoretical production of oxidating species due to a local criticality, corrosion penetration of neighbouring canisters will take in excess of 100 000 years, provided the reducing capacity of the tunnel filling is sufficient to take care of any oxygen arriving there.

Since the time that a chain reaction with significant energy generation can be sustained in the critical region is several orders of magnitude lower, it can be concluded that a local criticality does not significantly increase the risk of corrosion failure of nearby canisters in the repository.

FIGURE 1.



a) Velocity profile (Up) and concentration profile (c) in fissure around the clay filled hole.

Upstream deposition hole

Out diffusion of oxygen etc.

Downstream deposition hole

In diffusion of oxygen etc.

Appendix 5 Temperature perturbation in a spent fuel repository due to local criticality

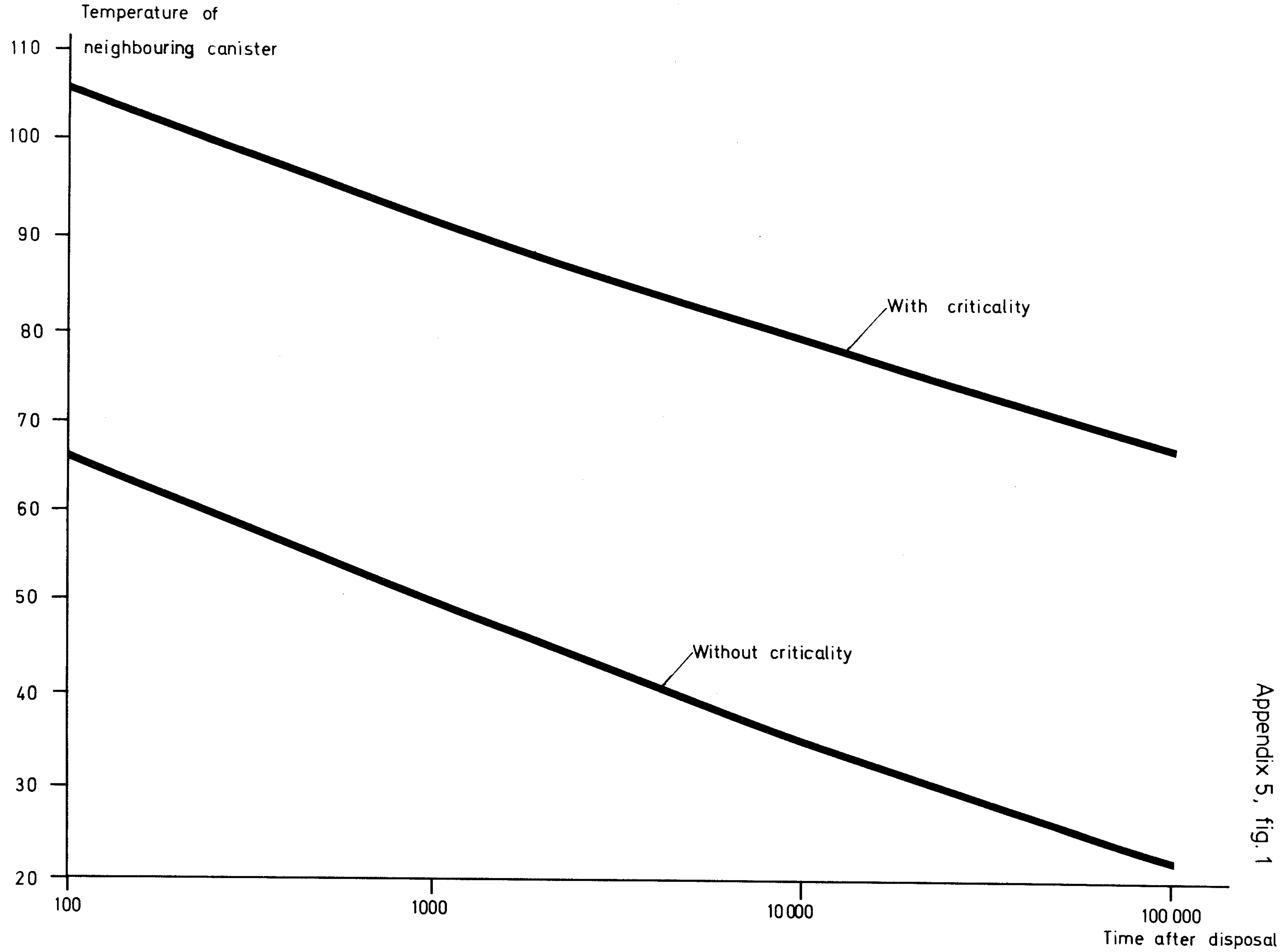
It is assumed that a local criticality has occurred in the immediate vicinity of a canister. The purpose of the calculation is to find the upper limit of the temperature increase in the neighbouring canisters caused by this.

The maximum temperature in the critical region is the saturation temperature of water at the hydrostatic pressure existing in the repository (about 5 MPa), i.e. 265°C. Because of the high conductivity of copper metal it was assumed that the whole defective copper canister is at this temperature.

The calculation were performed using a finite element three dimensional program and are described in detail in ref (14). The assumed temperature of the canister in the calcualtion was 300°C, i.e. slightly over-conservative.

Temperature calculations for a spent fuel repository without a local criticality have previously been performed. A comparison of the present calculations with these earlier values indicates a maximum temperature increase due to the criticality of about 45°C. The calculated temperature vs time curves with and without the temperature perturbation caused by the criticality are shown in figure 1.

The difference in the temperature curves seems considerable. However, sustaining the assumed canister temperature of 300°C requires an appreciable heat generation rate (reactor power), of the order of 10 kW. With this reactor power, the energy generation can only last for some 300 years because of the limitation in the amount of plutonium present. No appreciable consequences of such a short overtemperature episode can be envisaged.



Appendix 5, fig. 1

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COMPUTATION OF TEMPERATURE DISTRIBUTION IN THE FINAL STORAGE AROUND A CANISTER HEATED TO 300° C

Taivo Tarandi

Summary

Temperature distribution in the final storage of spent fuel due to local heating of a canister to 300° C has been computed for BWR-fuel.

The maximum temperature on the nearest neighborning canister is conservatively calculated to be 105° C, when the local heating up occurs 100 years after the deposition.

1. Introduction

There is a possibility of local temperature increase to 300° C in a canister [1].

The temperature distribution around such a canister is computed in the following for BWR-fuel.

The results are applicable also for PWR-fuel if the power per canister is the same.

2. Premises for calculation

The calculation is performed in the same way as the calculation of the temperature distribution in the storage without such defects [2].

The thermal conductivity of the bentonite is conservatively assumed to be 3 W/m° C.

With the aim of limiting the computer costs, the calculation is made in the form of stationary temperature distribution.

The assumption of constant generated power in the fuel leads to quite great conservatism in the beginning of the deposition as the power in reality sinks to a fraction of the initial value during the time to reach the steady state (in the order of thousands of years).

The calculation is made at following times and power densities.

Time after deposal years	Power per canister W
100	267,40
1 000	58,96
10 000	15,26

As a comparison an analogous steady state calculation is performed for the case without extremely heated canister.

3. Results

The results are shortly presented in Figures 1 - 3.

Fig. 1 shows the conditions when the heating of the canister starts at 100 years after the deposition. The temperature distribution as calculated for the actual power variation with time according to ref. [2], is also shown in this figure.

Thus, the temperature distribution calculated with constant power assumption, is much too conservative at this early time.

A more realistic estimation can be made in the following way.

The overtemperature in any point due to the heated canister is nearly proportional to the difference between the temperature of this canister and the background temperature in the storage. As this background temperature is lower at time 1 000 years according to fig. 2, we can assume that the overtemperature at the nearest neighbouring canister at 100 years is lower than corresponding overtemperature at 1 000 years. This leads to a maximum temperature of less than 105° C at the neighbouring canister. The corresponding temperature distribution is also shown in fig. 1.

At 1 000 years after the deposition, fig. 2, the steady state for the case without heated canister coincides with transient temperature distribution with actual power variation. Temperature increase 43° C due to heated canister is therefore correct at this moment.

At 10 000 years the corresponding increase is 45° C, which is somewhat higher due to the lower background temperature in the storage.

The sum up, the maximum temperature on the nearest neighbouring canister is as follows.

Time years	Max. temperature °C
100	<105
1 000	93
10 000	75

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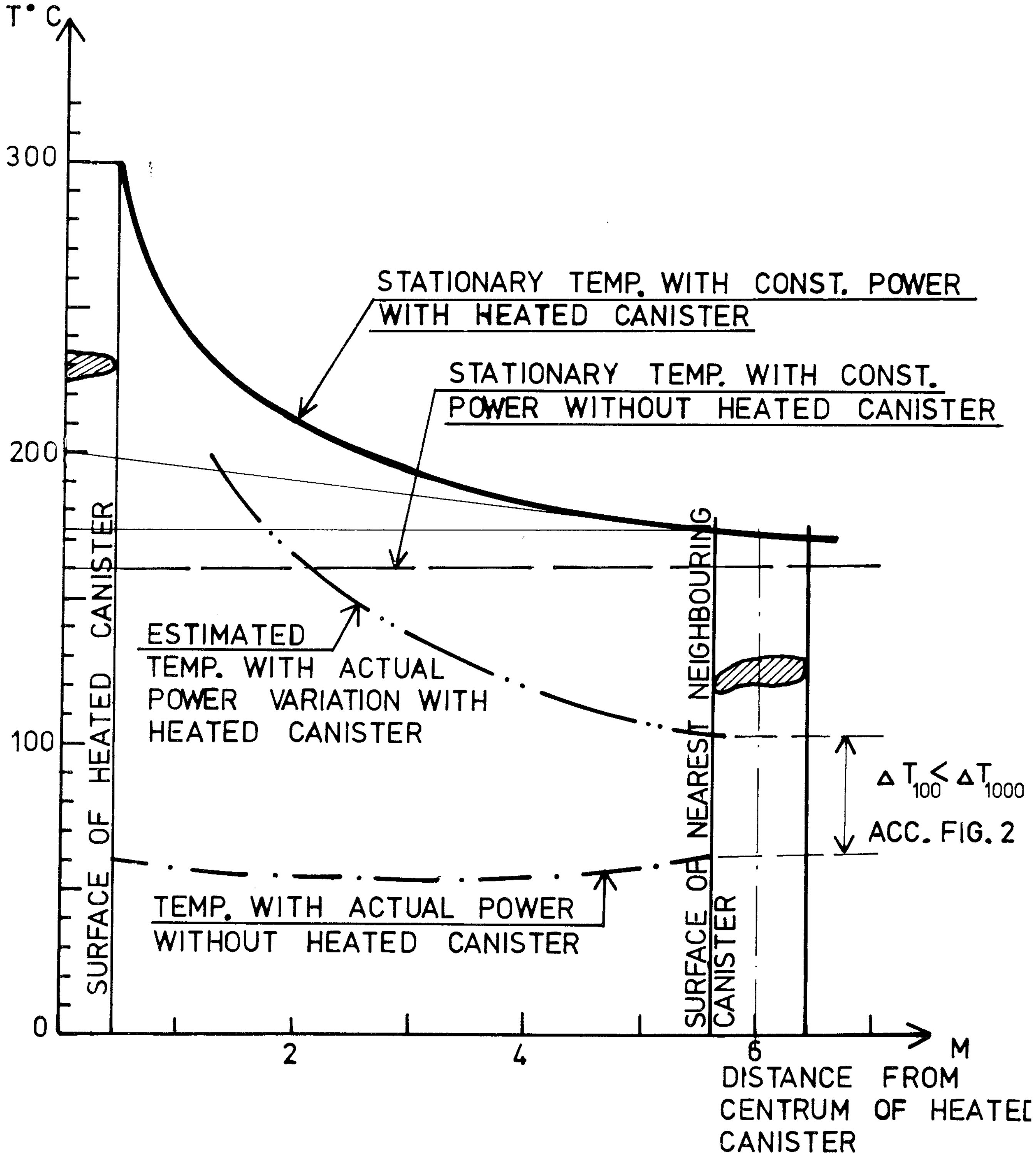

FIG.1. TEMPERATUREDISTRIBUTION
100 YEARS AFTER DEPOSITION

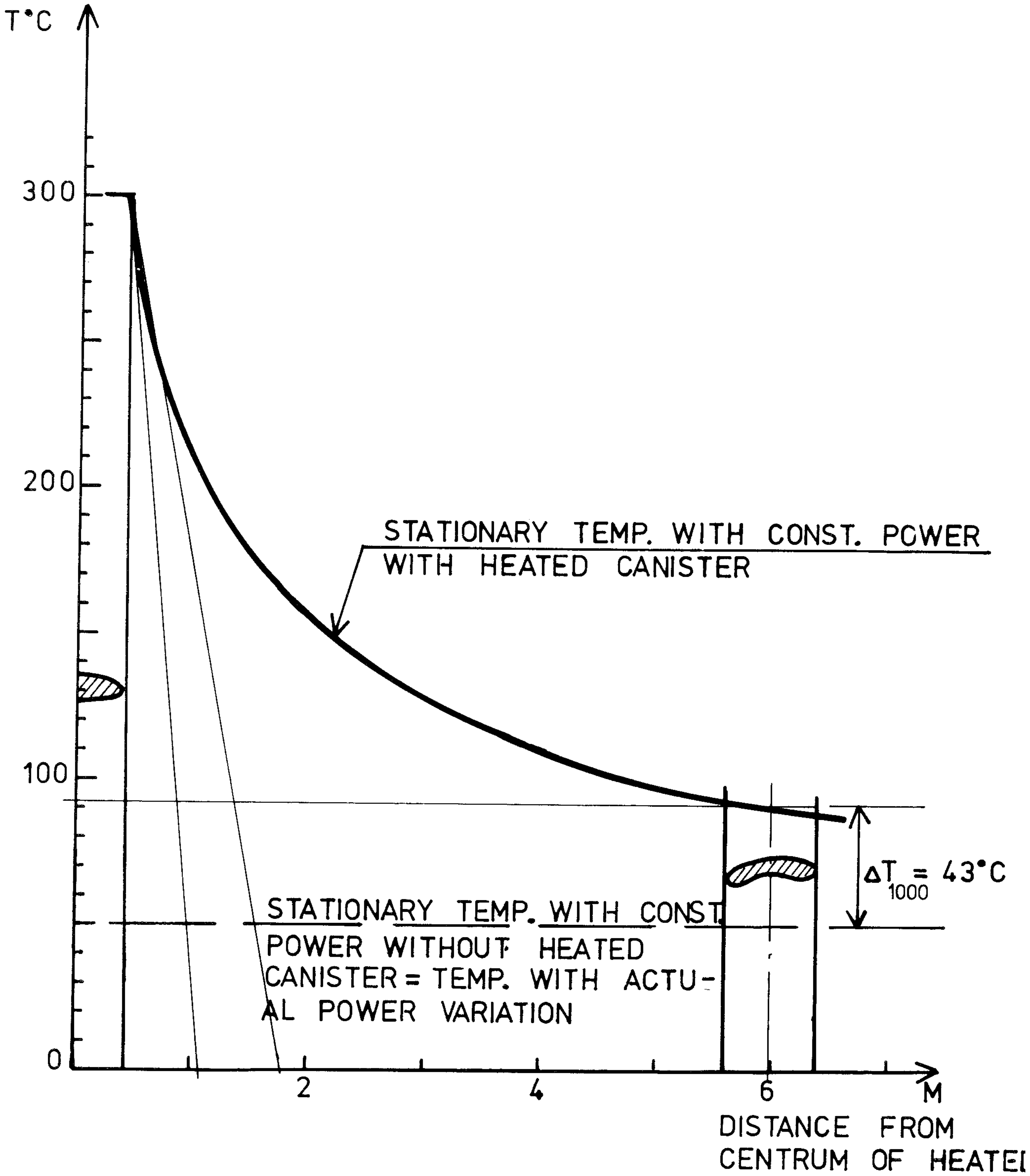

FIG. 2. TEMPERATUREDISTRIBUTION 1000 YEARS AFTER DEPOSITION

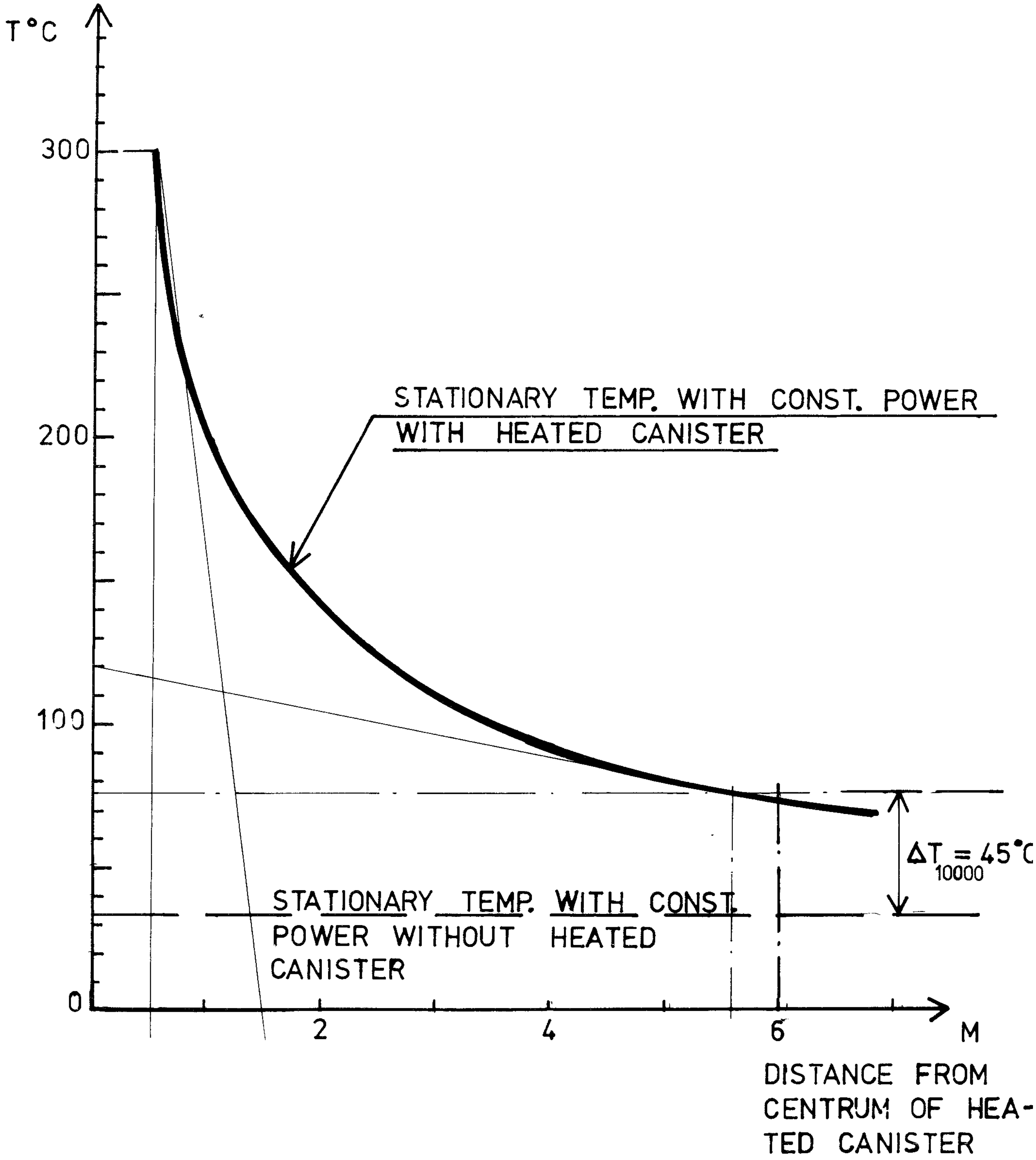

FIG. 3. TEMPERATUREDISTRIBUTION 10000 YEARS AFTER DEPOSITION

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