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# The Role of the Canister in a System for the Final Disposal of Spent Fuel or High-Level Waste

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# The role of the canister in a system for the final disposal of spent fuel or high-level waste

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A final repository for radioactive waste must isolate the toxic substances or distribute their release over time or space to avoid causing harmful concentrations of radionuclides in the biosphere.

The Swedish research has focused on a repository 500 m down in crystalline rock where the geochemical environment can give canisters a service life of the order of a million years. These evaluations are discussed and the safety effect of the canister is compared with that of other barriers available in a repository system.

Our conclusions are that a combined protection effect of natural and man-made barriers can be achieved that substantially exceeds what could reasonably be required by society. An actual repository design can then be based on an optimization of the cost to reach a level of accepted safety with due regard for the safety margins and redundancy necessary for achieving public confidence.

## 1. Background

The nuclear power programme in Sweden consists of 12 reactors at four sites with a total capacity of 9500 $MW_e$. According to Swedish legislation the owner of a nuclear power plant is responsible for the management of the radioactive wastes produced there. This responsibility is carried through the Swedish Nuclear Fuel and Waste Management Co. (SKB), an organization jointly owned by the nuclear power utilities in Sweden.

SKB is responsible for all handling, transportation and temporary as well as final storage of spent fuel or radioactive waste outside the power plants. The responsibility also entails the planning and construction of all the facilities and systems necessary for safe management, and all the research and development needed to achieve and prove an acceptable level of safety.

Since 1977, there is a special legal requirement concerning the radioactive waste that has had a major influence on the direction of the research and development effort in this field in Sweden. Before a new nuclear reactor is allowed to be taken into operation the owner has to prove, to the satisfaction of the Government, that the radioactive wastes produced by the operation of the reactor can be handled and finally disposed of in a safe manner.

As a consequence, the primary goal for the R&D has been to show the feasibility of safe disposal with no requirements for optimization.

The aim of the Swedish work has been:

(*a*) to evaluate the possible effects of available natural and man-made barriers in protecting man from the harmful effects of radioactive waste;

(*b*) to design a repository system that would give adequate long-term protection to man even under very pessimistic assumptions regarding the external circumstances and regarding man's ability to model relevant processes in Nature, and

(*c*) to show both that technology is available in Sweden for the construction of *man-made* barriers, and that sites exist in Sweden with *natural* barriers of such a quality that the total protection effect is acceptable to society.

The feasibility of a safe final storage has been evaluated and presented in two reports: one for the high-level waste from reprocessing of spent fuel (SKB 1977) and the other for the unreprocessed spent fuel as such (SKB 1983*a*). The Swedish Government has found that the feasibility of both methods was proved to a level acceptable to society.

This discussion of the role of the canister in a system for final storage of highly radioactive wastes is based on the Swedish work presented in the two reports mentioned above and will focus on four questions:

1. What effects do we want from a repository system?
2. What mechanisms or barriers do we have available?
3. What maximum guaranteed effectiveness can a canister be designed to?
4. What is the need for a long-lived canister in the total system?

## 2. Repository functions

If left uncontrolled, radioactive wastes can cause harm to man or his environment or both. The function of a final repository is to isolate the waste and allow for decay or to distribute the release over times long enough to dilute the waste so that man is not exposed to harmful concentrations of the nuclides. These repository functions can be provided by the near-field subsystem – consisting of the man-made barriers around the waste and also including those parts of the surrounding host rock whose natural characteristics have been significantly altered by the repository – or by the geosphere subsystem consisting of the natural bedrock along the pathways up to the biosphere. By repository design or by site selection both subsystems can be influenced by man. However, because a possible release from the repository to biosphere often is of much longer duration than the time needed for significant evolutionary change in the biosphere or long-term climatic changes, the biosphere is not included among the subsystems whose barrier effect can be influenced by man. In the discussions below the biosphere has been represented by a small lake recipient, with a slow turnover of water, often found in Swedish granite areas.

The need for isolation or distribution of the waste can be illustrated with a diagram for potential toxicity. In figure 1 the amount of each radionuclide remaining in 1 ton of spent fuel for different times after discharge has been multiplied by a factor giving the resulting effective dose commitment to an individual in the most exposed group if 1 Bq of that nuclide is released every year to the biosphere.

The nuclide content of the spent fuel as a function of time is typical of PWR fuel with a burnup of 38 $GW_d$/ton. The dose factors are taken from the KBS-3 report for a lake recipient and the most unfavourable pathway examined for each nuclide.

The diagram indicates which nuclides dominate the potential harm in a repository, and the relative importance of the radionuclides. The relative toxicity in the fuel during the first few hundred years is dominated by $^{137}Cs$. After that the actinides $^{241}Am$, $^{239}Pu$ and $^{237}Np$ dominate during a period up to 1 million years when the daughters of $^{238}U$ become dominant.

Since it is difficult to envisage a nuclide transport pathway to man shorter than 100 years from a deep geological repository on a selected site, and since an isolation of the waste for more than 1 million years would not greatly affect the potential toxicity of the waste, a meaningful


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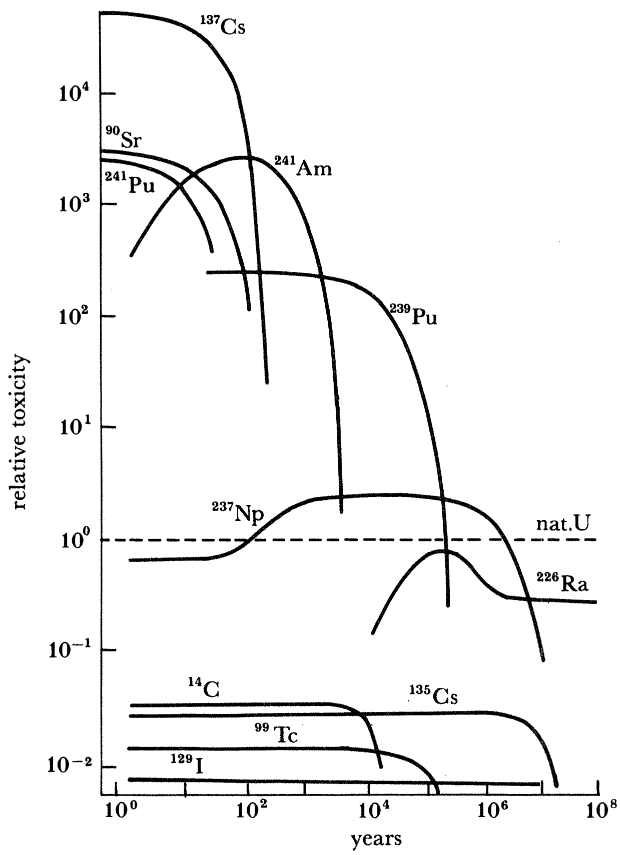


FIGURE 1. Potential toxicity of the radionuclides in spent nuclear fuel if released to the biosphere (each curve is plotted relative to the same mass of natural uranium).

target for the service life of a canister could be somewhere between 100 years and 1 million years.

At a given site the effect of a long isolation time can be replaced by a higher dilution in the biosphere by distributing the release over a longer time. It must be noted here, however, that the relative toxicity level marked with 1 is not an acceptability level. In fact even natural uranium in equilibrium with its daughters would have to be diluted considerably not to give unacceptable doses in the environment.

To give an illustration, the normal dilution in a typical small lake in Swedish granite sites would require the release of the 7000 tons of uranium in our repository concept to be distributed over 700000 years if it were not to give a dose above 0.1 mSv/year to the critical group.

## 3. THE REPOSITORY CONCEPT

To achieve the required isolation or distribution, natural barriers can be selected and man-made barriers can be designed to protect the waste.

The three main barriers we have investigated are:

(i) the canister isolating the waste from the groundwater;

(ii) the waste solubility limitations governed by the availability of groundwater and the chemical situation in the near field;



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(iii) the transport limitations for various radionuclides in the natural groundwaters of the surrounding bedrock including diffusion and sorption effects within the rock matrix.

Detailed investigations have been made for spent nuclear fuel but in principle the arguments are also valid for vitrified waste from reprocessing.

The following concept was developed. At 500 m depth in a selected host rock with low groundwater flow a tunnel system is excavated. Deposition holes are drilled in the tunnel floor,

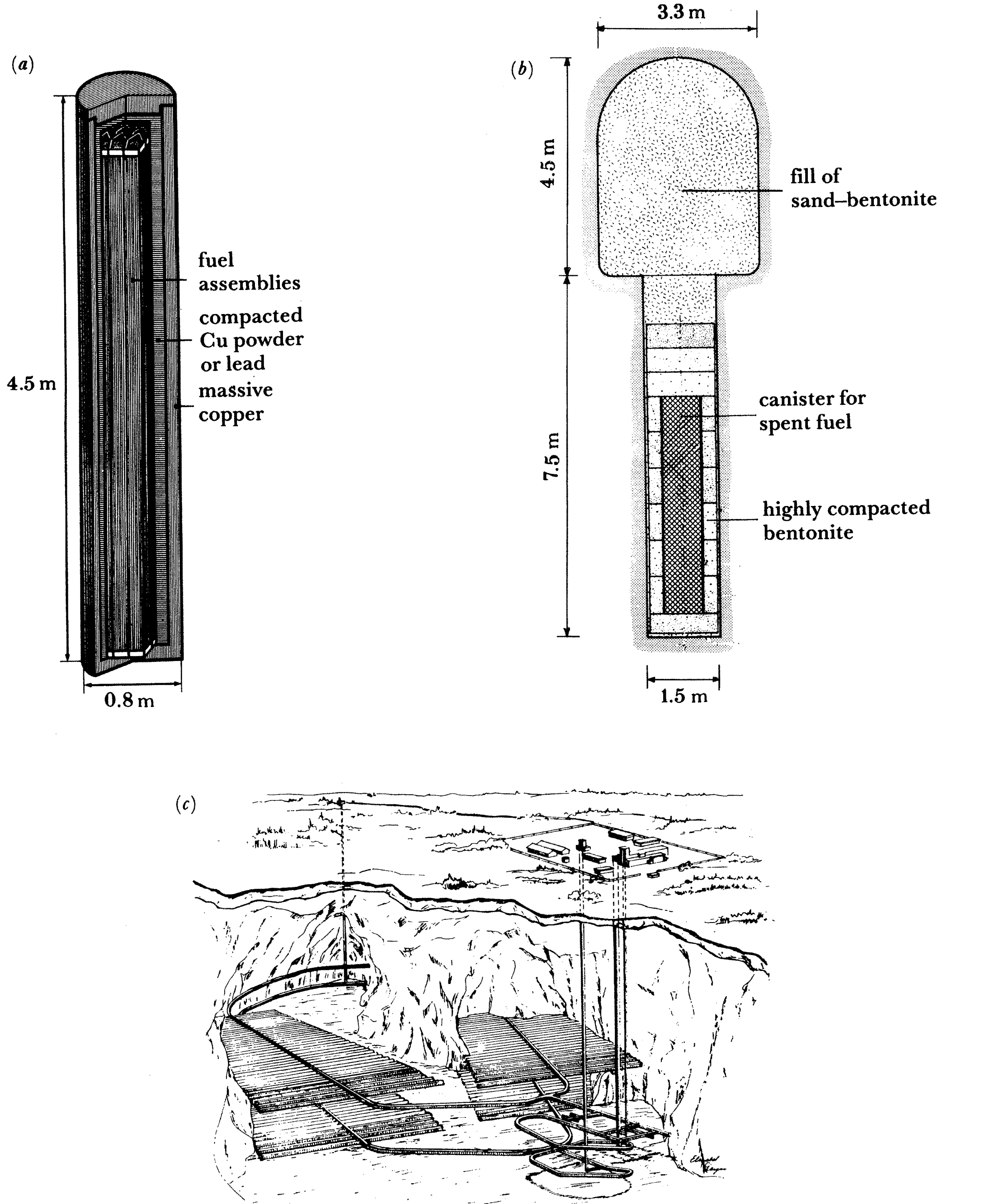


FIGURE 2. The Swedish KBS-3 repository concept. (*a*) Spent fuel in copper canister; (*b*) deposition hole with canister; (*c*) repository layout.

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and one canister with radioactive waste will be placed in each hole (see figure 2). The canister is made of copper 10 cm thick and contains the fuel assemblies. The free space left in the canister is filled to take the full pressure of the rock overburden. The space between the canister and the rock wall of the deposition hole is filled with compacted bentonite clay.

When the groundwater seeps into the repository, the clay will swell and become homogenized, thereby creating a zone around the canister which is impervious to water and permits mass transfer only by diffusion. This limits the inflow of corrosive substances from the groundwater to the canister surface, and enhances the canister life. At a later stage, when the canister is penetrated, the clay barrier will also limit the mass transport from the fuel to the flowing groundwater.

The quality of the final barrier, the geosphere surrounding the repository, can only be influenced by the site selection process. In our concept we have sought for areas with low groundwater flow surrounded by marked fissure zones.

## 4. The canister

A canister around the waste can have a number of functions:

(i) it serves as a handling container and radiation shield during preclosure operations;

(ii) it isolates the waste from groundwater contact during the initial period after closure when temperature and radioactivity are high and might influence the release of the radionuclides;

(iii) it can distribute the release from the total repository if the breakthrough time for the individual canister varies.

There are different ways to achieve a long service life for the canister. Either a thick wall is provided to compensate for the corrosion loss during the required service life, or a material is selected that is thermodynamically stable in the repository environment or protects itself with a corrosion-resisting surface layer.

In the feasibility study we selected a copper canister. The calculations of the service life of the canister are based on a maximum wall temperature below 100 °C and a groundwater chemistry found in Swedish bedrock at repository depth. Since copper is thermodynamically stable in pure water, corrosion is only possible by substances dissolved in the groundwater or left in the near-field after the deposition operations, for instance oxygen in the entrapped air or sulphides in the bentonite or the groundwater. There is also the possibility that oxidants formed close to the canister surface by gamma-radiolysis of water might have an effect on the corrosion rate.

Based on Swedish groundwater data and the repository concept presented above, the amount of copper that could become oxidized during different storage periods can be calculated (see table 1).

Table 1. Corrosion of copper by groundwater (SKBF/KBS 1983*b*)

| reactants | kilograms of corroded copper in $10^4$ years | $10^5$ years | $10^6$ years |
|---|---|---|---|
| oxidants formed by radiolysis | 0.4 | 1.1 | 8.8 |
| oxygen | 5.2 | 5.4 | 7.1 |
| sulphide | 6.2 | 8.3 | 29.0 |
| total | 12 | 15 | 45 |
| depth of deepest pit/mm | 3 | 5 | 17 |

To be able to estimate the service life of the canister the distribution of the corrosion over the container surface must be assessed. Based on corrosion studies of copper in soils, on archaeological artefacts of copper materials, and on investigations of native copper and of earth electrodes for lightning conductors, the corrosion experts in the Swedish study concluded that a pitting factor (the ratio between the depth of the deepest corrosion pit and the depth of the average corrosion), would probably be 5, and regarded 25 as a very pessimistic value. The last line of table 1 shows the depth of the deepest pit corroded in copper on the basis of a pitting factor of 25. Consequently, given the environmental conditions in possible Swedish repository sites a copper canister in a repository according to the KBS-3 concept has the capacity to isolate the waste from the groundwater over periods longer than 1 million years.

Such a statement is, of course, quite bold and is only valid if the assumptions made are valid over the same period of time. The very high geological stability of the Fenno-scandian shield and the fact that the hydrogeological régime found at our sites has been formed by a great number of successive glaciations has given us confidence to say that the next 1 million years, even including possible glaciations, will not drastically alter the present situation at a depth of 500 m. Since the bentonite clay is a volcanic ash clay, the proof of its stability over the 1 million year period is also based on geological evidence.

The degradation of the copper canister in the long term is governed by the amount of water that can exchange corrodants with the canister surface. Differences in water flow passing the deposition holes will also affect the canister break-through times. The repository in the Swedish concept covers an area of about 0.5 $km^2$ with 5000 canisters holding 1.4 t of uranium each. The investigations show that the sites have such a distribution of groundwater flow that if the thickness of the canisters is such that none will be penetrated before 100000 years in the most unfavourable part of the repository, the rest of the canister penetrations will be distributed over a period at least 10 times longer. This means that the canister will not only provide isolation but also distribute the release over a certain time period irrespective of other barriers.

The subsequent safety analysis in the KBS-3 report was based on the first canisters being penetrated at 100000 years and the rest distributed over the next 900000 years.

## 5. Other barriers

In the design and evaluation of the KBS-3 concept a number of additional barriers were studied.

The primary function of the bentonite clay around the canister is to limit the transport both of corrosive substances from the groundwater to the canister surface and of radionuclides from the fuel matrix to the groundwater. In our case the bentonite would restrict the exchange by a factor of about 3 compared with the canister in an empty water-filled hole. At a high groundwater-flow the effect is greater, at a small flow the effect is smaller. Other functions are to keep the canister centred, to protect it against smaller rock displacements, to act as a chemical buffer and to act as an ion exchanger.

The solubility limitation of the waste matrix does not provide an initial delay of the leaching process but distributes the release of radionuclides over time. An oxidizing zone is pessimistically assumed to be formed owing to alpha radiolysis at the fuel–water contact surface. Owing to the solubility of uranium in an oxidizing environment the dissolution rate will be about $2 \times 10^{-7}$/year, and those substances that are distributed in the uranium dioxide matrix will be released at the same rate if their own solubilities are not more limiting. Exceptions are carbon-14 in the fuel cladding, and the fraction of iodine and caesium that has accumulated in the pellet–cladding gap of the fuel rods.

As the migrating nuclides reach the natural iron(II)-rich granitic environment the redox potential will return to the natural reducing state. The lower solubility of uranium and neptunium in the reducing state will cause a precipitation to occur at the redox front. Calculations indicate that the distance of the redox front from the waste will be around 1 m. The further release of these nuclides will be governed by their solubilities in the reducing groundwater environment. The reduction of the release rate is for uranium about a factor 1000 and for neptunium about a factor of 3 compared with the release rate given by the dissolution of the matrix.

The final barrier is given by the geosphere around the site. Owing to sorption on fissure surfaces and diffusion into microfissures in the rock matrix, the radionuclides in the groundwater will move more slowly through the geosphere than the groundwater. Those substances that have a high tendency to sorb on the minerals available will be strongly delayed. Geotransport is the dominating factor for decreasing the release of $^{237}Np$ to the biosphere.

For the KBS-3 concept the barriers dominating the release limitation for the various substances in radioactive wastes are given in table 2.

Table 2. Barriers to release of radionuclides

(C, canister; M, matrix dissolution rate; O, solubility in oxidizing environment; R, solubility in the redox front; G, sorption in the geosphere.)

| | |
|---|---|
| carbon | C |
| iodine | M+C for 10% |
| caesium | M+C for 10% |
| technetium | R |
| uranium | R |
| neptunium | G (C) |
| plutonium | O |
| americium | G (C) |

To summarize the effects of the various barriers: (*a*) with the chemical conditions in granitic/gneissic sites in Sweden, the release of substances distributed in the waste matrix will be more strongly limited by solubility constraints than by the distribution of the canister failure over time; (*b*) substances in the fuel cladding or collected in the cladding gaps of the fuel rods are mainly limited by the canister.

If, however, a substantial fraction of a radionuclide is transported as a colloid or sorbed on other colloid particles the release rate based on solubility limitations at the waste surface or the redox front might be exceeded.

Furthermore, should sites with shorter pathlengths or quicker transport times up to the biosphere be selected, americium and neptunium release would be limited by the matrix solubility, or, for matrixes more soluble than the uranium dioxide fuel, by the canister.

Overall, then, given a high quality site with low groundwater flow, suitable chemistry and long and slow pathways up to the biosphere, repository concepts with also a quite limited isolation period given by the canister (at least down to 1000 years) could provide acceptable repositories. In fact a variation analysis in KBS-3 shows that the doses could be kept below 0.1 Sv/year also with initial defects in all canisters (figure 3).

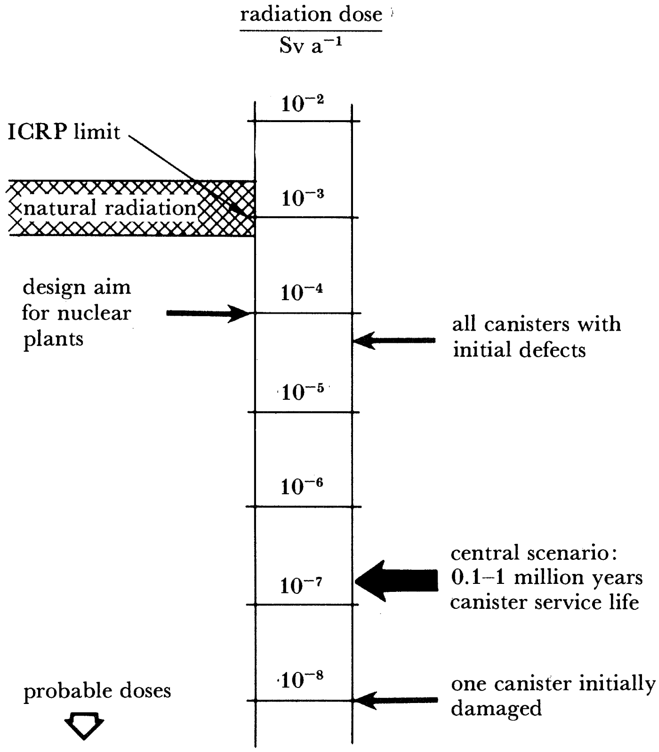


FIGURE 3. Calculated doses with and without a long-lived copper canister.

## 6. Cost and cost effectiveness

One of the main arguments against the use of copper in canisters has been the thought that the price of the copper would make the final repository system too expensive. Although the cost of copper in each canister is quite high (0.2 million SEK per canister with 10 cm wall thickness), in the KBS-3 concept the copper price will only form about 20% of the total cost for making a canister. Most of the cost is due to the need for radiological protection through radiation shielding and remote handling equipment, by the compartmentation and redundancies needed to achieve the required serviceability for the plant, and by the equipment and arrangements needed to make it possible to handle malfunctions and accidents in the plant (SBKF/KBS 1982).

Although this does not mean that substantial savings might not be made by reducing the wall thickness or by selecting other materials, the conclusion to be drawn is that cost does not prohibit the use of also a relatively expensive material in the canister, if there is a need for it in the system.

## 7. The role of long-lived canisters in a granite repository

In the Swedish study we concluded that there exist sites with good natural characteristics for a repository, and that we already have the technology to protect the environment from the waste by man-made barriers. Both these groups of barrier can be selected or made to such a high quality that the protection effect from the total system will substantially exceed what could reasonably be required by society.

The long-lived canister is thus not the sole solution to the problem of final storage of radioactive waste. On the contrary there seems to be a great potential to select both the number of barriers and their quality in such a way that the required level of protection will be achieved in a cost-effective way.

In this process of system optimization the availability of a long-lived canister might reduce the requirements for very small groundwater flow or a specific groundwater chemistry on a site. Another advantage in having an absolute barrier between the waste and the water during an early period is the simplification in the analysis that can be achieved. The initial period is avoided during which the radiotoxicity of the waste is highest, during which the temperature in the near-field of the waste is substantially elevated, and during which the chemical composition of the radioactive waste is most different from that of the analogue of natural uranium.

A final important role for the canister as well as for any of the other barriers, is to provide confidence in the safety of the repository even under unusual and extreme conditions. In a repository where multiple barriers, as independent in function as possible, provide the safety, the total safety of the repository will be less sensitive both to changes in environmental factors and to a possible lack of understanding of some interactions in the repository.

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