International Nuclear Fuel Cycle Evaluation

INFCE

INFCE/DEP./WG.4/63

PARTIAL PROCESSING

Co-Chairmen/WG4/54(A)
24th November 1978

Working Group 4: Reprocessing, Plutonium Handling, Recycle

Sub-Group 4A: Reprocessing

PARTIAL PROCESSING

Note by the Chairmen

In the work programme for INFCE Group 4A, we are due to consider the alternative reprocessing technologies as task 8. For that discussion, we have two papers from the Technical Secretariat (Co-Chairmen/WG4/18(A) and Co-Chairmen/WG4/52(A,B), a paper on co-processing from the Japanese delegation (Co-Chairmen/WG4/37(A)), and a paper on both co-processing and spiking from the American delegation (Co-Chairmen/WG4/38(A,B)). However, none of these papers cover the subject of partial reprocessing.

Paper Co-Chairmen/WG4/52(A,B) from the Technical Secretariat lists four definitions of terms which have already been agreed for use by Working Group 5. Partial processed material is there defined as:

> "A mixture or solution containing uranium and/or plutonium that is produced in a nuclear fuel processing plant by a process in which the fission products and/or the trans-plutonium actinides are not completely separated."

As Co-Chairmen, we have invited several delegations to present their thoughts on this subject but we have not, so far, identified a delegation willing to do so. The German delegation has now suggested that the most straightforward way of proceeding with this subject is for the co-chairmen to introduce the topic and he has stressed that he did not see this as in conflict with the Co-chairmen's role.

In response to this suggestion we now, with some diffidence, submit the attached paper written by one of us (WM). The paper describes the possible use of partial reprocessing in thermal reactor recycle, but suggests that there would be no merit in introducing it. We now wish to invite delegates to consider the arguments set out in that paper and, during the Vienna Meeting, we will consider whether those are sufficient to dismiss the subject from further consideration, or if not, what work we need to commission to assist our deliberations.

W. Marshall S. Tamiya

Co-Chairmen/WG4/54(A)

24th November 1978

THE USE OF PARTIAL PROCESSING IN THERMAL REACTOR RECYCLE

W. Marshall

One of the alternative reprocessing technologies which Working Group 4A needs to consider under Task 8 of its work programme is "partial processing". "Partial processed material" has been defined in Working Group 5 (as noted in Co-Chairmen/WG4/52(A,B))as:

> "A mixture or solution containing uranium and/or plutonium that is produced in a nuclear fuel processing plant by a process in which the fission products and/or the transplutonium actinides are not completely separated."

There are no doubt several methods by which it is technically feasible to modify a reprocessing plant flowsheet to permit some fission products and/or transplutonium actinides to be carried forward with the final uranium/plutonium product for incorporation in new fuel. This paper, which is based on a lecture delivered to the Uranium Institute earlier this year*, considers the possibility of applying to the recycle of plutonium in thermal reactors a particular method of partial processing which has been studied in collaboration with Dr. Chauncey Starr of the Electrical Power Research Institute in the USA together with our colleagues in EPRI and the United Kingdom Atomic Energy Authority. This method is based on the conventional PUREX process but has been named CIVEX to emphasise the differences. I must stress at the beginning that it was designed for use in a mature fast reactor fuel cycle (when fuel irradiated in a fast reactor would be reprocessed and fabricated into fresh fuel for a speedy return to a fast reactor i.e. 9-18 months) and not for use in thermal reactor recycle. It is, I believe, nevertheless a method on which Group 4A can reasonably base their discussion of partial processing since it allows me to set out the basic principles.

Figure 1, which shows the gamma activity of a spent thermal reactor fuel element, also shows beneath the main curve the contribution made to that activity by the various fission products present in it. Those curves illustrate how the activity is dominated in turn by short-lived fission products, a key group of isotopes with half-lives of several months, and a longer-lived group of fission products and actinides with half-lives of many years. The objective of partial processing would be to retain some of these fission products with the uranium and plutonium so that the level of radioactivity of the fresh mixed oxide fuel would be sufficient to prevent, or at least make very difficult, the illegal diversion of the plutonium for weapons purposes.

Figure 1 illustrates that if the fission products retained are to be sufficiently radioactive then the time at which reprocessing is carried out is of great importance. There is clearly a big difference between reprocessing such a spent fuel element within the first year, when the radioactivity of the fission products is very high, or reprocessing after 10 to 20 years when the radioactivity of the fission products has substantially decayed.

* "Nuclear Power and Non-Proliferation" by W. Marshall, Lecture to the Uranium Institute July 1978

In particular, it can be seen that if the fuel is reprocessed within the first year, then the radioactivity is dominated by the presence of several short-lived radioactive isotopes, namely zirconium, niobium, ruthenium, cerium and praesodymium whereas, after five years, these very active fission products have decayed and the radioactivity is then at a lower level and dominated by caesium, strontium and the actinides.

The CIVEX process is based primarily on the retention of the short-lived fission products named above. Table 1 shows the gamma activity and half-life for each of these isotopes and the final column gives a percentage of each which can be recycled together with the uranium and plutonium by suitable modifications of the reprocessing flowsheet. In a conventional reprocessing plant it is customary to include several stages of the plant to eliminate all the fission products from the final uranium and plutonium products. However, there is no technical reason why we are obliged to do this and, if the reprocessing flowsheet were simplified, then those fission products could be made to accompany the final plutonium-uranium product at any designed level. The plutonium would then be accompanied by such highly radioactive fission products that it would be virtually as "inaccessible" when it came out of the reprocessing plant as when it went into it.

The fabrication of this highly radioactive mixture into fresh fuel elements would certainly be more difficult than if the fission products had been separated out in the reprocessing plant. Nevertheless fabrication techniques could be developed, especially using the gel route, which has been under development for a number of years. The gel route fabricates fuel into perfect spherical particles which can then be loaded automatically and remotely into fuel pins. Once that process has been developed successfully for straightforward plutonium/uranium oxide fuel, there should not be great difficulties in extrapolating it to deal remotely with material containing fission products.

From the technical point of view, therefore, there seems no reason to suppose that the CIVEX process could not, in time, be successfully developed for use in thermal reactor recycle. Using the results in Table 1, the amount of activity which can be returned to thermal reactor fuel can be calculated. The results are plotted in Figures 2 and 3. The first curve in Figure 2 shows the activity of a thermal reactor fuel assembly as a function of time after removal from the reactor and the second curve shows the activity of the fresh thermal reactor fuel if prepared by a CIVEX process which recycles isotopes to the percentages described in Table 1. If the recycle time is shorter than or comparable to one year, then the activity of the fresh fuel is comparable to that of the old fuel. On the other hand, if the recycle time is longer, then the activity of the refabricated fuel will be smaller because the recycled fission products will have had time to decay. Figure 3 gives a percentage plot of the activity of the new fuel compared to the old.

These results do therefore show that it is possible to protect the plutonium in recycled thermal reactor fuel with activity to

roughly the same extent as it is protected in spent fuel discharged from a reactor, provided that such recycle is done rapidly i.e. before the recycled fission products have had time to decay.

There is in the world a very large surplus of unreprocessed spent fuel compared with reprocessing capacity and it seems unlikely that this situation will change very rapidly. The spent fuel which is reprocessed and any plutonium which is recycled in thermal reactors this century will therefore very largely be "old" spent fuel and 'old' plutonium, which may well be closely associated with long-lived actinides but cannot be associated with the fission products listed in Table 1 because they will have decayed. In practice it must be probable that ever into the next century most thermal reactor fuel will be reprocessed only after it has been in a cooling pond for a number of years. Finally, it must be recognised that it would be at least ten years before a CIVEX fuel cycle could be fully proven and available on a commercial scale.

In summary therefore this paper suggests that:

1. the recyle of fission products with uranium and plutonium in thermal reactor fuel would be technically feasible;

2. it would, however, take ten years or more to develop such a process to the point where it could be launched on a commercial scale;

3. since the majority of spent fuel to be reprocessed this century will have been in storage for ten years or more, the recycling of short-lived fission products with the U-Pu will not provide an effective means of making refabricated fuel "inaccessible" and therefore there is no merit in doing it.

TABLE 1

KEY FISSION PRODUCTS FOR "CIVEX" PROCESS

(450 kg assembly)

PWR Sub-assembly - 10 days After Discharge

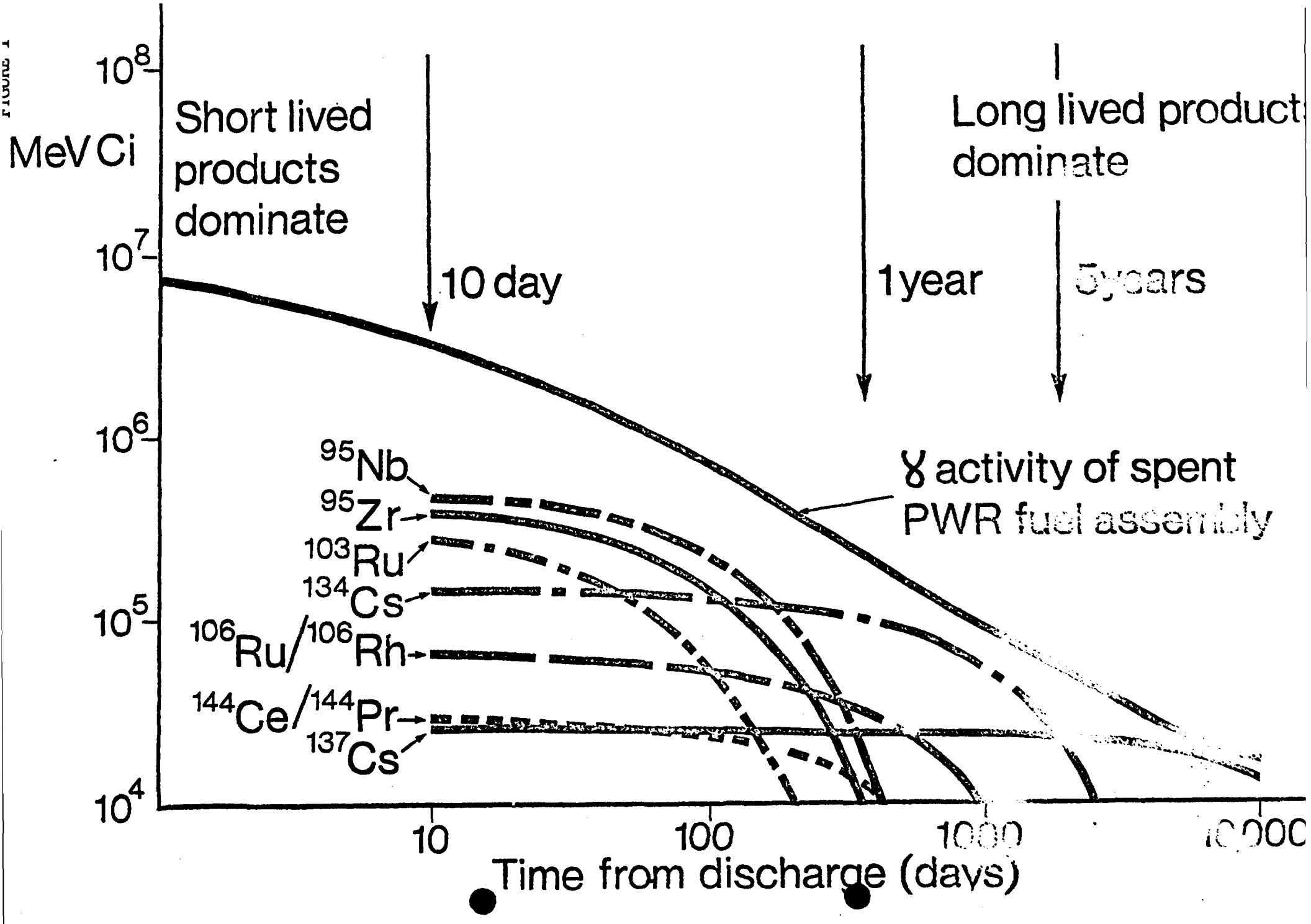
FISSION PRODUCT	ACTIVITY Ci MeV x 10^5	½ LIFE DAYS	PERCENTAGE RECYCLED
^{95}Nb	4.38	35	30
^{95}Zr	3.85	64	30
^{103}Ru	2.60	39	50
$^{106}Ru/^{106}Rh$	0.64	368	50
$^{144}Ce/^{144}Pr$	0.29	285	11

FIGURE LEGENDS

FIGURE 1 — The gamma activity of a spent PWR fuel assembly or a function of time and the contribution to that activity by several key isotopes.

FIGURE 2 — The gamma activity of a PWR fuel assembly and a new CIVEX assembly for a PWR,

FIGURE 3 — The ratio of the gamma activity of CIVEX fuel for a PWR to that of spent fuel from a PWR


MeV Ci
10^8
10^7
10^6
10^5
10^4
Short lived products dominate
10 day
1year
5years
Long lived product
dominate
γ activity of spent PWR fuel assembly
^{95}Nb
^{95}Zr
^{103}Ru
^{134}Cs
^{106}Ru/^{106}Rh
^{144}Ce/^{144}Pr
^{137}Cs
10
100
1000
10000
Time from discharge (days)

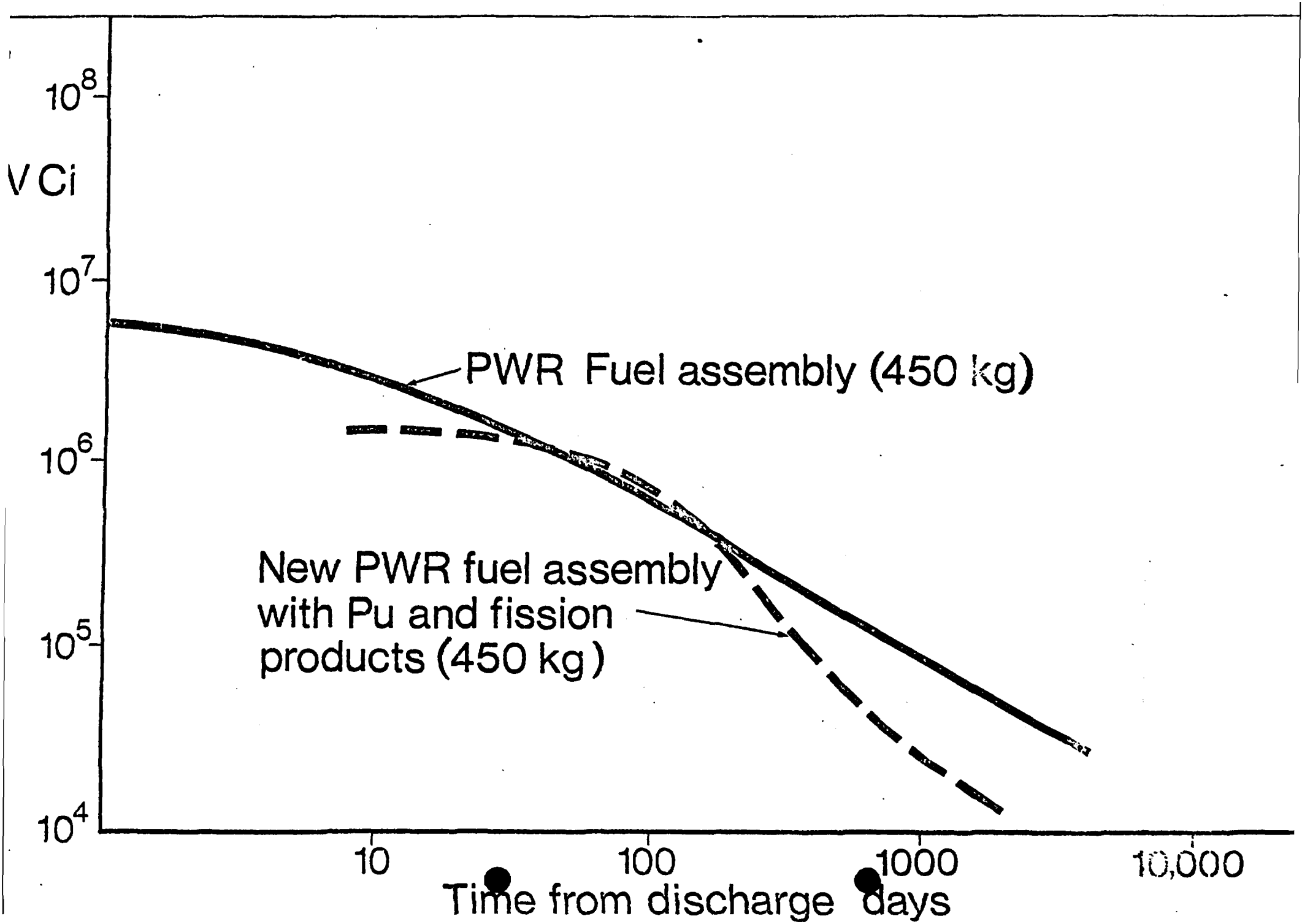
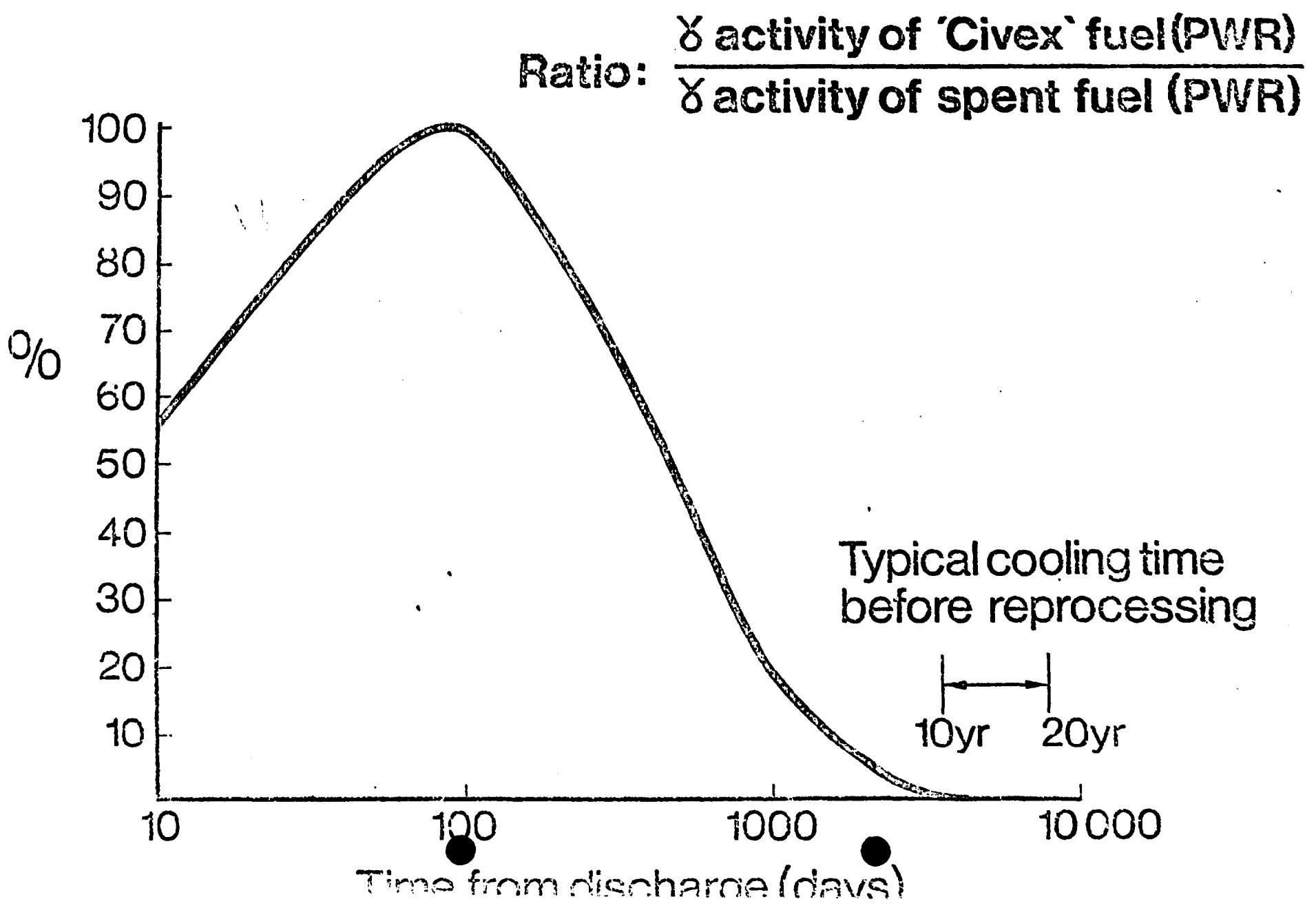

10^8
V Ci
10^7
10^6
10^5
10^4
PWR Fuel assembly (450 kg)
New PWR fuel assembly
with Pu and fission
products (450 kg)
10
100
1000
10,000
Time from discharge days

FIGURE 3


Ratio: γ activity of 'Civex' fuel (PWR) / γ activity of spent fuel (PWR)
%
100
90
80
70
60
50
40
30
20
10
10
100
1000
10 000
Time from discharge (days)
Typical cooling time before reprocessing
10yr
20yr