

# SKBF TECHNICAL KBS REPORT

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## The KBS UO<sub>2</sub> leaching program Summary Report 1983-02-01

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Nyköping, Sweden February 1983

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THE KBS UO<sub>2</sub> LEACHING PROGRAM

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This report concerns a study which was conducted for SKBF/KBS. The conclusions and viewpoints presented in the report are those of the author(s) and do not necessarily coincide with those of the client.

A list of other reports published in this series during 1983 is attached at the end of this report. Information on KBS technical reports from 1977-1978 (TR 121), 1979 (TR 79-28), 1980 (TR 80-26), 1981 (TR 81-17) and 1982 (TR 82-28) is available through SKBF/KBS.

**THE KBS UO<sub>2</sub> LEACHING PROGRAM**  
**Summary Report: 1983-02-01**

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The KBS UO<sub>2</sub> LEACHING PROGRAM  
SUMMARY REPORT: 1983-02-01

## ABSTRACT


The results so far obtained in the current KBS program on the leaching/corrosion of spent UO<sub>2</sub> fuel are presented and discussed.

Fuel with a burnup of about 40 MWd/kgU from a fuel rod from the Oskarshamn I BWR is used as source material.

Experiments have been performed mainly under oxidizing conditions, results being obtained which are in broad agreement with previously published work. An interesting feature of the experiments is the determination of the colloidal fraction of the material present in the aqueous phase after contact with the leachant. Such information, together with results of leaching at lower pH values, leads to the hypothesis that the concentrations of uranium and the activities in the aqueous phase are limited by solubility/adsorption effects.

A number of experiments have also been performed where preliminary attempts to impose the more realistic reducing conditions on the system have been made. Here, a small but significant reduction in the apparent leach rates has been achieved.

Approved by



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## A INTRODUCTION

This report presents the results at hand after one year of experimental work within the program. Results are available for contact times of up to 286 days in the sub-program where fuel specimens have been exposed to distilled water or synthetic ground water under oxidizing conditions at room temperature.

The pilot experiments performed in an attempt to attain reducing conditions are completed and are reported here, while the initial experiments to examine possible interaction effects between the fuel/water system and bentonite are still in progress.

## B LEACH SPECIMENS

A total of 38 segments of fuel and clad were cut from fuel rod A6 of fuel assembly 418 from the initial loading of the Oskarshamn 1 BWR. The rod had attained a mean burnup of 36.2 MWd/kgU and maximum burnup of 42 MWd/kgU. The latter value has been confirmed by burnup analysis of specimen 3.8 during the determination of fission product and actinide inventories, when a value of 41.4 MWd/kgU was obtained.

Fig 1 shows the positions of removal from the rod of the specimens for the various sub-programs, where experiments have in effect been performed in duplicate using specimens taken from different parts of the rod. It can be seen from the Cs-137 gamma scan of the rod also shown in the figure, that all specimens had a burnup of about 40 MWd/kgU.

## C LEACH PROGRAM

For a definition of program objectives and more detailed information on experimental procedure, the reader is referred to the previous Status Report NF(P)-82/49.

A schematic summary of the leach program is given in Fig 2, which shows that 4 sub-programs have been running concurrently:

- a) Oxidizing conditions (3A) In this sub-program the fuel/clad specimens - open-ended - are suspended by a Pt coil in 200 cm<sup>3</sup> of distilled water or a synthetic groundwater (for composition of the groundwater, see Table 1) in a 250 cm<sup>3</sup> Pyrex flask. The leachant is bubbled with pure N<sub>2</sub> gas prior to contacting with the specimen.

Experiment series 3.1 (distilled water) and 3.2 and 3.3 (groundwater) were leached according to a modified IAEA procedure, the same specimens being successively exposed to new leachant after each sampling time.

The fuel /clad specimens in the 3.4 and 3.5 series (except for an initial 14 day pre-leach to remove most of the gap inventory of the active cesium isotopes which otherwise would have further complicated the analytical procedures) were exposed statically to the leachant.

Fig 3 presents a sketch of the sampling procedure after each contact time.



After removal of the fuel specimen, the leachant is also removed from the flask, following which a 10 cm<sup>3</sup> aliquot is centrifuged through a so-called membrane filter (Aminco Corp, USA) whose 15-20 Å apertures are stated to retain material of molecular weight greater than 25 000. Both filtrate (sample A) and filter (sample C) are analysed for the species shown in the sketch.

After a rapid rinse of the empty flask to remove, hopefully, any micro-particles of fuel which may have fallen out from the specimens, the flask is exposed to 200 cm<sup>3</sup> of 5M HNO<sub>3</sub>/0.5M HF to dead-sorb any activity adsorbed on the vessel walls. After a few days, this solution (sample B) is also sent for analysis.

- b) Reducing conditions (3B) Six specimens, 3.6.1 - 3.6.3 and 3.7.1 - 3.7.3, were included in this sub-program. Each specimen was first pre-leached under oxidizing conditions for two periods of 91 days and 27 days respectively to remove most of the gap inventory of cesium activities. In the third stage of leaching, an attempt to impose reducing conditions on the new groundwater leachant was made by bubbling 6% H<sub>2</sub>/Ar through the leachant in the presence of a Pd catalyst deposited on a platinum mesh.

Three of the specimens (3.6.1.R; 3.6.2.R; 3.7.1.R) were then left in static con-

tact for 28 days, and one specimen (3.7.3.R) for 55 days. The remaining two specimens (3.6.3.R and 3.7.2.R) were exposed to a further treatment with 6% H<sub>2</sub>/Ar halfway through 56/55 day contact period.

At the conclusion of the leaching periods, all experiments in this series were sampled in the same way as for the oxidizing series.

- c) Bentonite interaction series (5) In this series, four fuel/clad specimens were preleached under oxidizing conditions for two periods of 91 and 27 days respectively and then contacted with new groundwater containing a suspension of crushed and sieved bentonite. Two of these latter experiments have been concluded, and analysis of the leachant and bentonite is in progress: The remaining two specimens are being exposed for longer contact times.
- d) Immersion tests (6) Two diametrical strips of fuel and clad have been cut out from the fuel rod (Specimens 6.1 and 6.2. See Fig 1.), ground and polished, and subjected to examination under the active optical microscope to characterize the microstructure of the fuel. For the second specimen, 6.2, a surface replica was also taken and examined by SEM.

Specimen 6.1 was then immersed in ground water under oxidizing conditions

for 6 months, while specimen 6.2 was immersed in groundwater treated with 6% H<sub>2</sub>/Ar in the presence of Pd catalyst in an attempt to impose reducing conditions. A six month exposure of this specimen is still in progress.

At the conclusion of the 6 month period, specimen 6.1 has been re-examined under the microscope to determine possible visible changes in the appearance of the fuel surface.

D           KBS UO<sub>2</sub> LEACH TESTS 1977 - 78

Prior to presentation of the results from the current program, it is interesting to review briefly those from the earlier, more limited program (TPM-BL-133 (REV)).

The 1977 - 78 program was restricted to 4 fuel/clad specimens of 18 - 19 mm in length, taken from fuel rod 391-F1 from the Oskarshamn I BWR, at two positions of different mean linear heat ratings and burnup, designated high and low power specimens respectively, as shown below:

	<u>kW/m</u>	<u>MWd/kgU</u>
High power specimens	24	26
Low power specimens	11	11.5

The fission gas release for the rod had been measured to be 0.11 % of the inventory.

The samples were suspended in stainless steel mesh baskets in 500 cm<sup>3</sup> of either distilled water or a synthetic groundwater similar to that used in the current program, but with about 300 ppm HCO<sub>3</sub><sup>-</sup> instead of 123 ppm. The pH was 8.5.

The tests were performed according to the modified IAEA procedure at 60°C up to a total contact time of only 105 days. After each contact time, the leachants were removed, acidified, and analyzed for cesium activities, Sr-90, uranium and total alpha activity. No attempt was made to determine the amount of possible colloidal material in the leachants, or on the vessel walls or the stainless steel baskets.

All the species measured showed much higher leach rates (expressed as fractions of the inventory leached per day) at the beginning of the tests, attributed in the case of cesium activities to rapid leaching of material present in or near the fuel/clad gap due to radial migration, and to the dissolution of fines present after sample cutting for the other species.

However, all values of leach rate decreased rapidly as a function of cumulative contact time and approached constant values at the conclusion of the tests after 105 days, being of the order of  $10^{-7}/d$  for total alpha activity, and  $10^{-6}/d$  for the other species.

The cumulative leach fractions after 105 days for Cs-137 were about 0.7 % and 0.03 % respectively, reflecting the strong dependence on linear heat rating of the well-established radial migration of cesium fission products.

More unexpectedly, a fourfold higher cumulative release of Sr-90 was found for the high power specimens compared with the low power specimens.

No differences in leach behaviour were found in the tests performed in distilled water and those in the synthetic groundwater, a surprising result in view of the well-known complexing of uranium with carbonate ion, and in conflict with some other published results.

## E CURRENT PROGRAM: RESULTS

The program so far has generated about 2500 separate analytical results. Due to the relatively large amounts of cesium activities leached from the fuel/clad gap during the initial leaching stages, it has often been found necessary to repeat the analysis of other gamma emitting nuclides after separation of the cesium activities, usually in the case of the filtrates after centrifugation. (Sample A: See Fig 3).

No such separation was required for any of the B samples (the 5M HNO<sub>3</sub>/0.5M HF solutions used to check the possible adsorption of activities on the vessel walls).

Even though in some cases the uranium contents in the B samples equalled or even exceeded those in the filtrates (A samples), calculation of the ratios to uranium of the measured fission product and actinide contents in all the 65 B solutions indicated, within the normal scatter associated with measurements at such low levels, that the ratios were characteristic of the original fuel inventories. Thus, the activity in the B solutions represented, as expected, the dissolution of microparticles of fuel which had fallen out from the suspended fuel specimens. In some B samples, uranium concentrations up to 1.5 ppm were detected, corresponding to the dissolution of only three fuel particles of radius 0.1 mm!

The important conclusion to be drawn from these results is that, even though it will be shown later in this report that colloidal forms of several of the fission products and actinides

are present in the leachant after contact with the fuel specimens, there is no evidence that significant quantities have been adsorbed on the vessel walls. Hence, it has been assumed in the following that the sum of the quantities determined in the A and C samples is a measure of the total release from the fuel to the leachant.

Because of the difficulty in defining the surface area of cracked  $UO_2$  fuel, the release fractions have been expressed as fractions of the initial inventories of fission products and actinides, all values being corrected for decay back to the reference date of 1982.02.14.

The individual inventories have been calculated from the known specimen weights, allowing for the weight of the Zircaloy clad, and the results of radiochemical analysis of reference specimen 3.8, slightly adjusted by the weighted means of the 65 B samples, which showed that there were small but significant differences between the actinide contents of the fuel in the sampling groups 3.3 - 5.1 and 3.1 - 3.8 respectively (see Fig 1.). These values of fission product and actinide inventories are considered to be accurate to within  $\pm 20\%$ .

The program results are presented in Tables 3 - 14. Since it will be postulated later in this report that substantial percentages of fission products other than cesium and strontium, and of the actinides are re-adsorbed on the fuel and/or clad surface after their initial entrance into the aqueous phase - presumably by congruent dissolution of the  $UO_2$  matrix - the terms Leach Fraction and Leach Rate have been avoided in the following.

Instead the terms FIAP (Fraction of Inventory in Aqueous Phase) and Apparent Leach Rate (FIAP/d) have been selected as representing better definitions of the quantities actually measured.

Also given in the tables are values of the contact times, incremental and cumulative, and pH, and the percentages of each measured species retained on the membrane filters after centrifugation.

As mentioned earlier in this report, the activity levels of the fission products Ru-106, Ce-144 and Eu-154 were below the levels of detection in some samples. In those cases where only a result from an A or a C sample was available, the mean values of the percentages retained on the membrane filters over the whole program for these nuclides have been used to estimate (A + C) values. The apparent leach rates calculated in this way are given in parenthesis in the tables.

Even the uranium concentrations were often close to detection levels and the analytical results were expressed as "less than" values. These limits have, of necessity, been viewed as measured values, which represents conservatism, but which causes greater scatter in the results for uranium than for other species.



## F DISCUSSION

F.1 Sample contamination problems

As can be seen from the schematic summary of the leach program shown in Fig 2, a number of blank runs, usually with distilled water, were also incorporated in the program. This was felt to be essential in view of the large number of sample solutions and sampling and replacement operations within the confined limits of the concrete cell.

These solutions were removed at intervals and sampled, centrifuged etc according to the same procedures employed for the active runs in order to assess the risks of radioactive cross-contamination. Analysis of the filtrates and membrane filters from these blank runs suggested that such problems of cross-contamination were negligible.

However, another unanticipated and still not satisfactorily explained contamination incident occurred during the program which perturbed the pattern of results but which, in retrospect, was fortunate for their evaluation. Also shown in Fig 2 are the numbers of the separately - prepared batches of groundwater (G1, G2 etc) introduced into the cell and the values of pH measured on completion of the contact periods. In August 1982 it was found that almost all samples of groundwater of batch G3 and a distilled water sample which were introduced into the cell during the preceeding May, showed low pH values.

Formation of nitric acid in the nitrogen saturated solutions due to radiation was suspected but thought unlikely in view of the fact that the static tests 3.4.4 and 3.5.4 removed at the same time showed no pH decrease in spite of larger radiation doses.

Samples of the three solutions which had shown the largest pH decreases and others which had shown no pH change were analyzed by ion chromatography. The analytical results, which are given in Table 2, show the presence of fluoride in the three low pH solutions, and in one of them - 3.2.5 - even a significant nitrate content. The nitrate/fluoride ratio in this sample was similar to that in the 5M HNO<sub>3</sub>/0.5M HF deadsorption solution, strongly suggesting the contamination with this solution of most of the leachants introduced into the cell during May 1982.

As a consequence of this, the on-going incremental leach tests 3.1.6 and 3.2.6, with new batches of distilled and groundwater respectively, were terminated after 104 days and analyzed. The measured pH values were found to be normal, and the apparent leach rates, which had been high in the low pH samples, returned to low values.

F.2 Oxidizing conditions

The data referring to oxidizing conditions is extensive, since in addition to the results from the 3A program itself, additional data is available from the pre-leaching phases of the 3B and 5 series.

The cumulative FIAP values (in %) for Cs-137 for the 3A series are presented in Fig 4. The values for series 3.1, 3.2 and 3.5 and for series 3.3 and 3.4 are plotted separately to show the small scatter in the data and to illustrate the significant difference between the two groups, reflecting the positions in the rod from which the fuel/clad specimens were taken. The amount of cesium migrating radially through the fuel to the vicinity of the fuel/clad gap is dependent on the irradiation history of the fuel at that position.

It can be seen from Fig 4 and the data tables that 0.7 - 1% of the total Cs-137 inventory is readily accessible to the leachant and is removed within the first week, corresponding to mean ALR values of over  $10^{-3}$ /d. (It should be recalled that the measured release of Kr and Xe isotopes from the fuel in the whole rod during operation was 0.7% of the inventory).

The values of ALR then decrease rapidly, leveling off to approach values of  $3 - 4 \cdot 10^{-6}$ /d. (The abrupt rise in cumulative FIAP for series 3.2 shown in Fig 4A correspond to the specimen of groundwater with pH 2.3: a significant increase in matrix dissolution occurred in this leachant. See Table 4).

The results for the static test series agree well with the corresponding IAEA type tests.

From Fig 4A, it appears that higher leachability was obtained in distilled water but this may be a fortuitous result depending on the sample position in the rod.

The results for Cs-134 are not presented in this report since they were essentially identical with those for Cs-137.

The cumulative FIAP for Sr-90 are plotted in Fig 5 against cumulative contact time on a log/log scale, the results again being grouped according to specimen position in the fuel rod. A slope of +1 on such a log/log plot implies a constant apparent leach rate, and lines corresponding to ALR values of  $10^{-5}$  and  $3 \cdot 10^{-6}/d$  have been included in the figure to illustrate the trend. If the Cs-137 results can be considered representative mainly of its dissolution in or near the fuel/clad gap, the hypothesis is advanced here that the behaviour of Sr-90 is characteristic, perhaps even most characteristic, of congruent fuel matrix dissolution.

For the first group of specimens (Series 3.1, 3.2 and 3.5) initial values of ALR over  $10^{-5}/d$ , were obtained but decreasing with cumulative contact time. (Note that the results for the IAEA type series include the "spuriously" high results for the low pH contact periods between 91 - 182 days, apparently increasing the slope).

Initial values of ALR were lower for the second group (3.3 and 3.4 series) with as yet no apparent trend towards decreasing values.

Summarizing, the apparent leach rate for Sr-90 in the high burnup fuel used in the program is about  $3 - 4 \cdot 10^{-6}/d$  after 200 days of cumulative contact time, (about the same as for Cs-137), somewhat higher than in the earlier KBS program, and over an order of magnitude higher than the value obtained for a similar leachant by Johnson (1) in leach tests on CANDU fuel.

In this report the term "colloidal" has been defined arbitrarily as the material retained on the membrane filter after centrifugation of the leachant. It is estimated that about 5% of the  $10 \text{ cm}^3$  sample aliquot is retained within the pores of the filter, thus giving a bias to the values of colloidal fraction of this size.

Inspection of the data tables then leads to the conclusion that with one exception, series 3.1 with distilled water leachant, only negligible quantities of both Cs-137 and Sr-90 were present in the leachants as colloids. In distilled water, however, a value of about 15% for Cs-137 and values over 50% for Sr-90 were obtained.

The behaviour of uranium and the transuranides on leaching is best discussed against the background of the cumulative FIAP values for the IAEA-type series given in Figs 6-8, again presented as log/log plots. It is seen that the lines for uranium lie about one order of magnitude, and for plutonium, americium and curium about two orders of magnitude lower than the corresponding Sr-90 lines in all samples prior to the low pH contact periods mentioned previously.

The effect of contact with leachant of low pH, however, is seen to be striking, giving large

step increases in the cumulative FIAP values, reflecting the values of ALR extracted from tables 3-5 and reproduced below:

Leachant	<u>ALR (FIAP/d x10<sup>6</sup>)</u>			
	3.2.4	3.1.5	3.2.5	3.3.5
pH	8.3	3.9	2.3	4.8
Sr-90	2.7	5.8	14	3.6
U	0.12	3.6	12	0.56
Pu-239/240	0.02	0.18	1.5	0.20
Pu-238/Am-241	0.02	1.2	2.7	0.45
Cm-242/-244	0.01	8.7	11	2.3
Ru-106	-	1.1	11	(0.45)
Ce-144	-	5.7	11	2.4
Eu-154	0.03	3.4	11	1.7

Compared with the results for leachant 3.2.4, which had a pH of 8.3, all apparent leach rates increase as a function of decreasing pH, the largest increases being for the curium isotopes which have about the same ALR as Sr-90 between pH 2.3-4.8. Uranium attained the same ALR as Sr-90 only at pH 2.3, while plutonium still had a significantly lower value.

The ALR values for Pu-238/Am-241 appear to be intermediate between those for Pu-239/-240 and the curium isotopes.

However, the analytical method used for the transuranides was alpha spectrometry which gives a peak at 5.5 MeV with contributions from both Pu-238 and Am-241 which cannot be resolved. The

Am-241 contribution to the joint peak is calculated by the BEGAFIP code to be about 15%, and using this figure, it can be shown that the ALR values for the joint peak represent the means of individual values for Pu-238 behaving like Pu-239/-240 and Am-241 behaving like the curium isotopes.

From Tables 3 - 5 it can be seen that the lanthanide fission products Ce-144 and Eu-154 behave similarly to curium.

On examining the values of the percentages retained on the membrane filters, the following general conclusions can be drawn:

- at values of pH 7 - 8.5, a substantial part (> 50%) of the released uranium and plutonium was in the colloidal form during the first few sampling stages but then decreased to low values.
- at all values of pH, most of the curium was in the colloidal form, usually 90 - 100%.
- at low pH, virtually no uranium or plutonium was in the colloidal form.

The results from the static tests (series 3.4 and 3.5), which are presented in Tables 6 - 9, and plotted as cumulative FIAP values against cumulative contact time in Figs 9 - 12, essentially confirm the results discussed above for the IAEA-type tests. One difference, however, should be noted: after the longest static contact times - 182 days - the quantities of uranium and transuranides in the aqueous phase were

less than in the preceeding 91 days contact periods suggesting either saturation or read-sorption effects.

In comparison of the results of the current program with those from the earlier KBS leach tests, and from the Canadian studies reported by Johnson (1) and Vandergraaf (2), a number of similarities and differences can be noted.

The initial rapid leaching of cesium, corresponding to the dissolution of cesium compounds, perhaps cesium uranates, in the vicinity of the fuel/clad gap is well established. The fraction of the local cesium inventories in the fuel leached during this phase (weeks) is of the same order as the release of the inert gases krypton and xenon from the fuel at the same position during operation and is correlated with the local irradiation history.

However, the results from both Canadian test series suggested a preferential long-term leaching of cesium at contact times up to 800 days. In the experiments reported here, apparent leach rates for Cs-137 and Sr-90 were similar after contact times of 200 - 300 days. (See results for samples 3.1.6, 3.2.6 and 3.3.5).

The major, and as yet unexplained, anomaly in the results concerns the behaviour of uranium: in the Canadian work, carbonate-containing groundwaters were found to give leach rates an order of magnitude higher than distilled water, and only slightly lower than the values for Sr-90, whereas in both KBS series, no differences in uranium behaviour in these leachants was observed.



The results from all the experimental programs are in general agreement with respect to the fission products and the other actinides, since it has always been observed that the fractions of their inventories in the aqueous phase correspond to apparent leach rates ranging from  $10^{-6}/d$  to  $10^{-8}/d$ , with Sr-90 lying highest and plutonium towards the lower value.

It is in the context of this spread in apparent leach rates that the results from the inadvertent low pH leach periods have been extremely useful. Inspection of the table of ALR values on page 17, shows that already at pH 4.8, the apparent leach rates for the lanthanides cerium and europium, and curium (and americium: see above) approach the value for Sr-90 (which is still roughly the same as for high pH exposures) while the values for uranium and plutonium have increased but not to the same extent.

This trend is continued for sample 3.1.5 (pH 3.9) where only plutonium still has an ALR significantly lower than Sr-90. Finally, in sample 3.2.5 (pH 2.3) it is seen that all species with the exception of plutonium have, within experimental error, attained the same value as Sr-90.

Low pH leaching is also seen to be accompanied by a decrease in the colloid fraction for curium in groundwater, while no retention on the membrane filters was observed for either uranium or plutonium.

On the basis of the above observations the following description of the dissolution process

and explanation of the observed spread in apparent leach rates can be advanced.

At a pH of 2.3, the fuel matrix dissolves at a rate of about  $10^{-5}$ /d, all species with the exception of plutonium being held in the aqueous phase, although the lanthanides and trivalent actinides are partially (20 - 30%) in the colloidal form. About 90% of the plutonium, however, is readsorbed on the fuel/clad surface.

At pH 4, the rate of matrix dissolution decreases by about a factor of 2, and even dissolved uranium begins to be adsorbed on the surface. Finally, somewhere about pH 5, the amounts of the lanthanides and trivalent actinides retained in the aqueous phase are strongly reduced by the same surface adsorption phenomenon.

Thus the lower values of apparent leach rates for uranium, the transuranides and the lanthanides are postulated to be due to their different surface adsorption properties, and it is suggested here that the measured leach rates for Sr-90 probably represent the best measure of the fuel matrix dissolution.

It should be noted that such adsorption behaviour is expected from the known solution chemistry of these elements, and is supported by Johnson's results (1) on the adsorption of radioactive tracer Ce-144 on unirradiated  $UO_2$  and clad, finding over 90% adsorbed after 7 days contact time in a carbonate-containing groundwater.

### F.3 Reducing conditions

The results for the pilot experiments under reducing conditions are presented in Tables 10 - 12. In each of the six experiments, the first of the two pre-leaches were performed in groundwater with inadvertently low pH (~5) and it can be seen that the results are in general agreement with the discussion under F.2 above.

However, the purpose of the tests was to examine the potential of the H<sub>2</sub>/Pd system as a means of attaining reducing conditions in the leachant, and the experimental limitations of performing the experiments in glass apparatus in an air-atmosphere concrete cell.

Inspection of the results designated R, the reduction stage, and with the assumption that the Sr-90 leach rate is a good measure of matrix dissolution, shows that in four of the samples a reduction in dissolution rate by a factor of about 30 was indeed attained, while one sample showed some reduction, and one none at all.

Other interesting observations are that all species except cesium and strontium were present in the aqueous phase mainly in the colloidal form, and that in two of the specimens, enhanced ruthenium dissolution is suggested.

Obviously, these results, while fairly satisfactory, fall short of demonstrating the very large reduction in dissolution rate under reducing conditions that has been suggested on the basis of thermodynamic calculations. Nor do they shed any light on the possible limitation of such a potential reduction due to local oxidative

dissolution of the matrix caused by alpha radiolysis in a thin leachant film at the fuel surface.

Further experiments will be performed in this series.

#### F.4 Bentonite interaction series

As mentioned previously, analytical work is still in progress. However, the results from the oxidative pre-leach tests are shown to support the hypothesis advanced in F.2.

F.5 Immersion tests

So far only specimen 6.1, that immersed under oxidizing conditions, has been examined. It had been feared that the specimen surface would have been, at best, partially obscured by debris from the plastic/graphite specimen mount, but it was found that the surface was in good condition.

Examination under the active optical microscope showed that the surface was populated by particles which appeared to be amorphous, or at least could not be clearly resolved, ranging in size between 3 - 20  $\mu\text{m}$ . No apparent variation in population density over the specimen diameter was observed.

After examination under the optical microscope, a replica of the surface was made which was then examined in the scanning electron microscope. (The specimen itself was too active to permit direct examination). It had been hoped that at least some of the particles would have accompanied the replica, enabling a closer examination of their nature. It was found, however, that the replica was free of particles, possibly due to dissolution in the methyl acetate used in stripping the film from the specimen or by physical agitation. A further examination of the specimen surface showed that they were no longer present.

Assuming that the particles were spherical and consisted of  $\text{UO}_3 \cdot 0.5 \text{H}_2\text{O}$ , a very rough calculation showed that this would represent a surface concentration of 10 - 20  $\mu\text{g U/cm}^2$ . This is an order of magnitude less than the amount

calculated to be present if the following assumptions are made:

- a leach rate for uranium of  $3 \cdot 10^{-6}$ /d over the 180 day contact period
- 90% of the uranium is deposited on the surface
- a value for the specific surface area of the  $UO_2$  fuel of  $2 \text{ cm}^2/\text{g } UO_2$ . (Johnson's value: ref 1).

The value of 90% for adsorption on the surface, based on the results of leaching of fuel with adherent clad thus restricting leachant flow, may well be too high for the freely exposed specimen 6.1. Also, the value of  $2 \text{ cm}^2/\text{g}$  for the surface area of cracked spent fuel is certainly conservatively low. More realistic values for these two assumptions would appreciably increase agreement between the calculations.

Thus, although the results are far from conclusive, there seems to be some evidence supporting the hypothesis of major re-adsorption effects advanced earlier in this report.

Specimen 6.1 was also examined very carefully before and after immersion in an attempt to establish if dissolution of the fuel matrix occurs essentially at the grain boundaries.

Even here the results are inconclusive since grain boundaries are difficult to see in the unetched condition, the contact time was only 180 days, and slight differences in surface

illumination and in photographic recording of the results can affect the conclusions.

Close inspection of the microphotographs indicated the presence of new etch pits, sometimes in the form of thin channels, at the grain boundaries. However, it seems that longer immersion times are necessary in order to firmly establish this observation.

## G CONCLUSIONS

The experimental results presented in this report are in general agreement with those in other published work.

The release to the leachant, at an early stage, of the so-called gap inventory of those fission products which are very mobile in the  $UO_2$  lattice - cesium, which was measured in this program, but also probably iodine and tellurium - is well established, as is its quantitative relationship to the release of the inert gases to the gap and plenum during operation.

Under oxidizing conditions, apparent leach rates for Sr-90 in both distilled water and groundwater of about  $3 \cdot 10^{-6}/d$  were obtained for the high burnup fuel (40 MWd/kgU) used in this program. This value, which is higher than published values for fuel of lower burnup, is expected to decrease as a function of increasing contact time.

With some reservations for preferential leaching of possible strontium - rich inclusions in the fuel, the hypothesis is advanced here that the Sr-90 results best represent the dissolution of the fuel matrix itself. The lanthanide fission products and the actinides, including uranium, are thought to be retained in the aqueous phase after initial dissolution only in small amounts, most being readsorbed on the surface of the fuel and/or clad.

Evidence of such readsorption effects include the observation of (unidentified) particles on the fuel surface after immersion tests, but



mainly variation of the measured apparent leach rates as a function of leachant pH.

At low pH (2.3) the dissolution of the  $UO_2$  matrix increases by about a factor of 5 compared with that at pH 7 - 8.5. Even at this pH, plutonium is still largely reabsorbed on the surface, while the other species are retained in the aqueous phase.

Pilot tests using the  $H_2/Pd$  system as a means of imposing reducing conditions on the leachant gave encouraging results. A decrease in the dissolution rate of the fuel matrix, as measured by the Sr-90 leach rate, by about a factor of 30 was obtained, which is somewhat higher than that obtained by Norris (3) under glove-box conditions.

## H ACKNOWLEDGEMENTS

It is a pleasure to acknowledge the leadership of Dr Lars Werme, the KBS project leader, during the inception and course of this program. Thanks are also due to many within Studsvik Energiteknik AB who have contributed to the program, in particular Kerstin Svanberg, Arne Holmér and Birgit Bivered of the Section for Fuel Performance, and Yvonne Sandell and Hans Tovedal of the Analytical Section.

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Table 1. Composition of the synthetic ground-water.

Species	Molarity	ppm
$\text{HCO}_3^-$	2.014 $10^{-3}$	123
$\text{SiO}_2$	2.056 $10^{-4}$	12
$\text{SO}_4^{2-}$	1.000 $10^{-4}$	9.6
$\text{Cl}^-$	1.973 $10^{-3}$	70
$\text{Ca}^{2+}$	4.477 $10^{-4}$	18
$\text{Mg}^{2+}$	1.774 $10^{-4}$	4.3
$\text{K}^+$	1.000 $10^{-4}$	3.9
$\text{Na}^+$	2.836 $10^{-4}$	65

pH: 8.0 - 8.2

Ionic strength: 0.0085

Table 2. Post-leach analysis results on selected leach solutions.

Leachant	Cl <sup>-</sup> (ppm)	F <sup>-</sup> (ppm)	NO <sub>3</sub> <sup>-</sup> (ppm)	pH
3.1.4	<2	<1	<20	7.0
3.1.5	<2	5	<20	3.9
3.2.5	130, 115	23	850, 720	2.3
3.3.4	72	<1	<10	8.3
3.3.5	68, 88	12	<20	4.8
3.4.5	72	<1	<20	8.0
3.5.4	75	<1	<20	8.4
3.5.5	24	<1	<20	8.4

SAMPLE	3.1.1		3.1.2		3.1.3		3.1.4		3.1.5		3.1.6	
CONTACT TIME (DAYS)	7		7		14		63		91		104	
CUMULATIVE CONTACT TIME (DAYS)	7		14		28		91		182		286	
P <sub>H</sub>	N.M. (~ 7)				7.3		7.0		3.9		7.5	
	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%
Cs-137	1425	15	95	18	56	26	12	13	6.8	6	0.73	18
Sr-90	15	53	8.3	64	4.8	74	3.6	53	5.8	30	1.4	78
Ru-106	0.29	43	(0.06)		-		-		1.1	4	(0.02)	
Ce-144	(0.06)		(0.05)		-		-		5.7	94	(0.005)	
Eu-154	(0.06)		-		-		-		3.4	94	(0.09)	
U	1.3	29	1.0	22	0.11	50	0.02	55	3.6	4	0.02	80
Pu-239/-240	0.40	42	0.51	18	0.16	15	0.09	17	0.18	0	0.18	22
Pu-238/Am-241	0.26	27	0.49	19	0.13	13	0.07	8	1.2	86	0.15	20
Cm-242/-244	0.13	91	0.22	91	0.02	95	0.01	84	8.7	96	0.08	99

Table 3. 3.1 Series (Modified IAEA method: Distilled water). Values of Apparent Leach Rate (FIAP/d x 10<sup>-6</sup>) and percentages retained on membrane filter.

SAMPLE	3.2.1		3.2.2		3.2.3		3.2.4		3.2.5		3.2.6	
CONTACT TIME (DAYS)	7		7		14		63		91		104	
CUMULATIVE CONTACT TIME (DAYS)	7		14		28		91		182		286	
P <sub>H</sub>	N.M. (~ 8.2 - 9)				8.1		8.3		2.3		8.3	
	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%
Cs-137	1130	10	62	5	26	10	10	4	16	3	1.2	7
Sr-90	23	23	11	18	12	12	2.7	7	14	4	0.5	6
Ru-106	0.70	26	0.07	40	-		-		11	2	0.05	35
Ce-144	(0.07)		(0.02)		-		-		11	25	-	
Eu-154	-		(0.04)		(0.04)		0.03	27	11	23	(0.006)	
U	1.1	9	0.3	67	0.44		0.12	9	12	2	0.12	17
Pu-239/-240	0.13	53	0.04	73	0.22	19	0.02	12	1.5	0	0.004	31
Pu-238/Am-241	0.10	53	0.03	73	0.20	9	0.02	9	2.7	32	0.003	31
Cm-242/-244	0.10	100	0.03	100	0.04	73	0.01	46	11	37	0.003	86

Table 4. 3.2 Series (Modified IAEA method: Groundwater). Values of Apparent Leach Rate (FIAP/d x 10<sup>-6</sup>) and percentages retained on membrane filter.

SAMPLE	3.3.1		3.3.2		3.3.3		3.3.4		3.3.5	
CONTACT TIME (DAYS)	7		7		14		63		91	
CUMULATIVE CONTACT TIME (DAYS)	7		14		28		91		182	
P <sub>H</sub>	N.M. (~ 8.2 - 9)				8.1		8.3		4.8	
	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%
Cs-137	1332	9	187	6	60	6	15	4	2.4	4
Sr-90	6.5	15	4.5	15	12	7	5.3	7	3.6	7
Ru-106	1.1	40	0.18	41	0.06	8	-		(0.45)	
Ce-144	(0.11)		(0.04)		-		(0.03)		2.4	39
Eu-154	(0.21)		(0.06)		-		-		1.7	35
U	1.5	25	0.37	53	1.4	7	0.21	11	0.56	5
Pu-239/-240	0.42	60	0.10	73	0.39	7	0.04	8	0.20	0
Pu-238/Am-241	0.32	62	0.10	81	0.35	6	0.04	9	0.45	39
Cm-242/-244	0.32	97	0.08	100	0.02	88	0.003	88	2.3	51

Table 5. 3.3 Series (Modified IAEA method: Groundwater). Values of Apparent Leach Rate (FIAP/d x 10<sup>-6</sup>) and percentages retained on membrane filter.



SAMPLE	3.4.1		3.4.2		3.4.3		3.4.4		3.4.5	
CONTACT TIME (DAYS)	7		14		14		77		168	
CUMULATIVE CONTACT TIME (DAYS)	7		14		28		91		182	
P <sub>H</sub>	N.M. (~ 8.2 - 9)				8.2		8.3		8.0	
	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%
Cs-137	1388	6	829	9	4.7	7	22	7	12	3
Sr-90	3.8	12	3.4	14	8.1	7	4.1	11	4.0	7
Ru-106	1.0	28	0.57	34	0.05	21	-	-	-	-
Ce-144	(0.11)		(0.06)		(0.01)		-		-	
Eu-154	(0.14)		(0.10)		(0.05)		-		(0.01)	
U	1.6	59	0.91	22	1.4		0.34	5	0.08	6
Pu-239/-240	0.18	58	0.27	64	0.39	10	0.07	15	0.008	8
Pu-238/Am-241	0.23	78	0.16	47	0.36	9	0.07	16	0.007	8
Cm-242/-244	0.36	79	0.29	100	0.08	94	0.009	93	0.0007	75

Table 6. 3.4 Series (Static method: Groundwater). Values of Apparent Leach Rate (FIAP/d x 10<sup>-6</sup>) and percentages retained on membrane filter.

SAMPLE	3.4.3.14		3.4.4.14		3.4.5.14		3.4.6.14		3.4.7.14		3.4.8.14	
CONTACT TIME (DAYS)	14		14		14		14		14		14	
CUMULATIVE CONTACT TIME (DAYS)	14		14		14		14		14		14	
P <sub>H</sub>	N.M. (~ 8.2 - 9)											
	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%
Cs-137	816	4	862	5	876	5	831	4	683	5	755	3
Sr-90	4.1	16	3.4	17	3.0	17	3.1	16	6.5	10	3.8	14
Ru-106	0.68	24	0.61	32	0.67	32	0.80	27	1.0	25	0.91	26
Ce-144	(0.26)		(0.16)		(0.11)		(0.01)		0.12	86	(0.09)	
Eu-154	(0.28)		(0.24)		(0.17)		(0.18)		0.20	53	(0.21)	
U	0.63	8	0.64	15	0.4	35	0.52	9	1.3	11	0.84	18
Pu-239/-240	0.23	33	0.23	44	0.26	30	0.34	37	0.59	12	0.30	27
Pu-238/Am-241	0.20	48	0.22	58	0.24	47	0.29	47	0.52	21	0.25	39
Cm-242/-244	0.26	100	0.28	100	0.37	100	0.47	100	0.47	81	0.32	100

Table 7. 3.4 Series (Continued). Values of ALR and percentages on membrane filter for pre-leach samples.

SAMPLE	3.5.1		3.5.2		3.5.3		3.5.4		3.5.5	
CONTACT TIME (DAYS)	7		14		14		77		168	
CUMULATIVE CONTACT TIME (DAYS)	7		14		28		91		182	
P <sub>H</sub>	N.M. (~ 8.2 - 9)				8.2		8.4		8.4	
	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%
Cs-137	1269	7	631	4	50	23	11	4	4.6	5
Sr-90	13.9	15	13.3	15	16.8	13	5.5	7	2.7	8
Ru-106	0.64	23	0.26	19	-		-		-	
Ce-144	(0.006)		(0.10)		-		-		-	
Eu-154	(0.05)						0.05	35	(0.003)	
U	1.0	19	0.26		0.76	20	0.16	6	0.03	50
Pu-239/-240	0.09	59	0.05	50	0.25	17	0.06	6	0.005	56
Pu-238/Am-241	0.11	63	0.04	56	0.23	11	0.08	5	0.005	36
Cm-242/-244	0.10	98	0.03	100	0.04	95	0.02	42	0.0005	29

Table 8. 3.5 Series (Static method: Groundwater). Values of Apparent Leach Rate (FIAP/d x 10<sup>-6</sup>) and percentages retained on membrane filter. .

SAMPLE	3.5.3.14		3.5.4.14		3.5.5.14		3.5.6.14		3.5.7.14		3.5.8.14	
CONTACT TIME (DAYS)	14		14		14		14		14		14	
CUMULATIVE CONTACT TIME (DAYS)	14		14		14		14		14		14	
P <sub>H</sub>	N.M. (~ 8.2 - 9)											
	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%
Cs-137	558	5	645	5	560	5	509	5	532	4	581	4
Sr-90	13.6	14	13.3	18	10.4	18	6.6	20	3.3	15	4.4	16
Ru-106	(0.31)		(0.23)		(0.41)		(0.35)		(0.37)		(0.43)	
Ce-144	-		-		-		-		-		-	
Eu-154	-		-		-		-		-		-	
U	0.71	14	0.74	20	0.29	50	0.29	50	0.32	50	0.49	60
Pu-239/-240	0.06	69	0.04	54	0.06	46	0.04	68	0.02	55	0.05	80
Pu-238/Am-241	0.05	57	0.04	49	0.05	43	0.03	71	0.02	65	0.03	81
Cm-242/-244	0.03	98	0.02	100	0.03	100	0.02	100	0.02	100	0.03	100

Table 9. 3.5 Series (Continued). Values of ALR and percentages on membrane filter for pre-leach samples.

SAMPLE	3.6.1.1		3.6.1.2		3.6.1.R		3.6.2.1		3.6.2.2		3.6.2.R	
	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%
CONTACT TIME (DAYS)	91		27		28		91		27		28	
CUMULATIVE CONTACT TIME (DAYS)	91		118		146		91		118		146	
P <sub>H</sub>	4.7		8.1		8.6		4.6		8.2		8.5	
Cs-137	107	4	3.3	3	3.3	4	95	3	3.1	3	2.9	4
Sr-90	7.2	6	3.0	6	0.13	18	7.6	5	2.7	7	0.10	9
Ru-106	0.19	6	0.05	34	0.53	43	0.54	3	0.05	30	0.57	88
Ce-144	0.71	77	-		0.03	92	1.7	22	(0.007)		0.04	83
Eu-154	0.66	62	(0.02)		0.02	71	0.60	33	(0.04)		0.07	65
U	0.59	2	0.53	15	0.03	77	0.54	3	0.47	17	0.04	80
Pu-239/-240	0.04	0	0.11	13	0.05	97	0.06	0	0.09	13	0.07	86
Pu-238/Am-241	0.18	48	0.11	10	0.06	97	0.15	37	0.08	12	0.06	84
Cm-242/-244	1.4	45	0.02	84	0.06	91	1.1	38	0.02	76	0.12	69

Table 10. 3.6/3.7 Series (Static method: Groundwater. Oxidizing pre-leach/reducing tests). Values of Apparent Leach Rate (FIAP/d x 10<sup>-6</sup>) and percentages retained on membrane filter.

SAMPLE	3.6.3.1		3.6.3.2		3.6.3.R		3.7.1.1		3.7.1.2		3.7.1.R	
	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%
CONTACT TIME (DAYS)	91		27		56		91		27		28	
CUMULATIVE CONTACT TIME (DAYS)	91		118		174		91		118		146	
P <sub>H</sub>	5.0		8.4		8.9		4.7		8.6		8.5	
Cs-137	95	3	2.9	3	2.7	4	94	5	3.1	3	60	3
Sr-90	6.9	5	2.7	6	4.9	0	6.8	8	2.2	5	0.73	3
Ru-106	0.38	3	0.05	23	0.09	98	0.60	5	0.04	36	0.01	78
Ce-144	0.68	38	-		(0.01)		1.7	54	(0.005)		-	
Eu-154	0.60	35	(0.02)		0.01	81	1.4	47	-		-	
U	0.39	3	0.62	21	0.08	78	0.78	10	0.49	13	0.09	100
Pu-239/-240	0.02	0	0.08	0	0.01	89	0.33	21	0.09	12	0.003	78
Pu-238/Am-241	0.15	39	0.09	7	0.01	99	0.64	40	0.08	9	0.002	77
Cm-242/-244	1.2	36	0.02	74	0.03	97	2.7	46	0.01	85	0.002	86

Table 11. 3.6/3.7 Series (Continued).

SAMPLE	3.7.2.1		3.7.2.2		3.7.2.R		3.7.3.1		3.7.3.2		3.7.3.R	
CONTACT TIME (DAYS)	91		28		55		91		28		55	
CUMULATIVE CONTACT TIME (DAYS)	91		119		174		91		119		174	
P <sub>H</sub>	4.5		8.0		8.9		5.0		7.9		8.8	
	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%
Cs-137	91	3	3.1	4	4.8	3	92	4	2.5	4	41	3
Sr-90	7.0	5	2.4	7	0.11	7	6.5	6	2.0	9	0.09	6
Ru-106	0.74	4	0.03	32	0.006	70	0.52	3	0.03	36	0.002	48
Ce-144	1.3	25	-		(0.002)		1.3	41	(0.004)		-	
Eu-154	1.6	22	(0.03)		(0.005)		1.3	28	(0.02)		-	
U	0.73	6	0.80	9	0.08	85	0.52	7	0.54	18	0.08	83
Pu-239/-240	0.09	0	0.08	11	0.005	100	0.05	0	0.06	13	0.0008	57
Pu-238/Am-241	0.37	41	0.09	10	0.006	93	0.36	44	0.06	8	0.0005	67
Cm-242/-244	2.3	35	0.009	91	0.004	98	1.6	6	0.009	79	0.0006	73

Table 12. 3.6/3.7 Series (Continued).

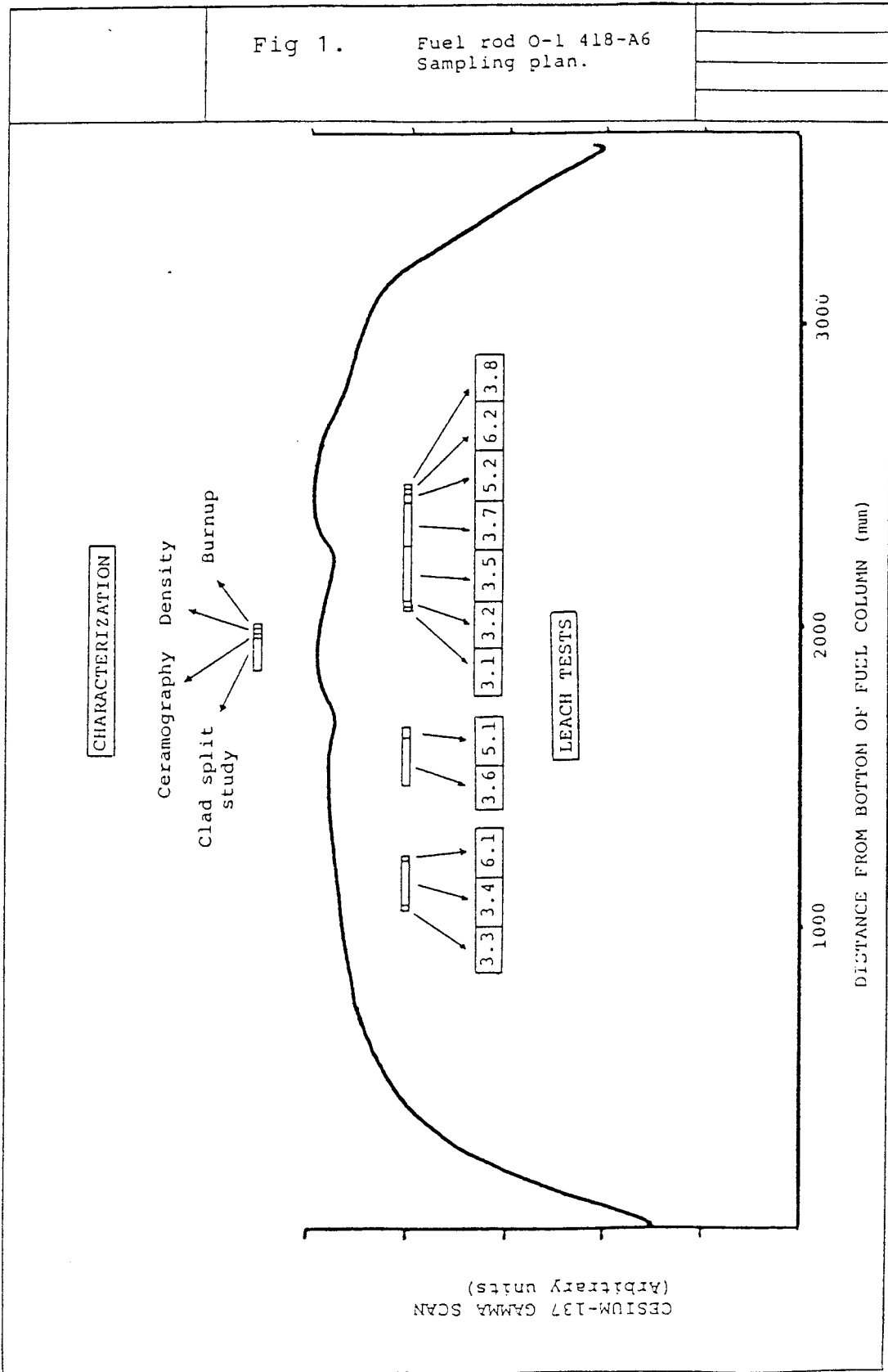
SAMPLE	5.1.4.1		5.1.4.2		5.1.4.B		5.1.7.1		5.1.7.2		5.1.7.B	
	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%
CONTACT TIME (DAYS)	91		27				91		27			
CUMULATIVE CONTACT TIME (DAYS)	91		118				91		118			
P <sub>H</sub>	5.1		8.1				5.6		8.1			
Cs-137	132	2	4.8	3	I N P R O G R E S S		113	3	4.6	4	I N P R O G R E S S	
Sr-90	6.6	4	4.1	8			5.5	5	4.2	8		
Ru-106	0.24	3	(0.05)				0.17	5	(0.05)			
Ce-144	0.39	37	(0.01)				0.07	49	-			
Eu-154	0.36	30	(0.02)				0.09	40	(0.01)			
U	0.16	10	0.36	29			0.34	11	0.76	14		
Pu-239/-240	-	-	0.09	6			0.005	100	0.19	5		
Pu-238/Am-241	0.09	29	0.09	6			0.02	52	0.19	5		
Cm-242/-244	0.66	36	0.02	74			0.12	45	0.02	85		

Table 13. 5.1/5.2 Series (Static method: Groundwater. Pre-leach/Bentonite interaction tests). Values of Apparent Leach Rate (FIAP/d x 10<sup>-6</sup>) and percentages retained on membrane filter.



SAMPLE	5.2.4.1		5.2.4.2		5.2.4.B		5.2.7.1		5.2.7.2		5.2.7.B	
	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%	ALR	%
CONTACT TIME (DAYS)	91		27				91		27			
CUMULATIVE CONTACT TIME (DAYS)	91		118				91		118			
P <sub>H</sub>	5.3		8.2				8.2		8.2			
Cs-137	101	3	4.2	3			101	3	3.0	3		
Sr-90	6.4	5	1.9	7	I N P R O G R E S S		6.7	7	2.7	3	I N P R O G R E S S	
Ru-106	0.36	4	(0.05)				(0.02)		(0.02)			
Ce-144	0.79	35	-				(0.003)		-			
Eu-154	0.88	32	(0.02)				(0.02)		(0.05)			
U	0.29	8	0.56	14			0.15	11	0.30	53		
Pu-239/-240	-	-	0.11	10			0.04	10	0.04	9		
Pu-238/Am-241	0.29	39	0.10	8			0.05	13	0.04	9		
Cm-242/-244	1.4	34	0.02	79	0.01	78	0.04	78				

Table 14. 5.1/5.2 Series (Continued).



32 0700 1114

4-91  
10-13552

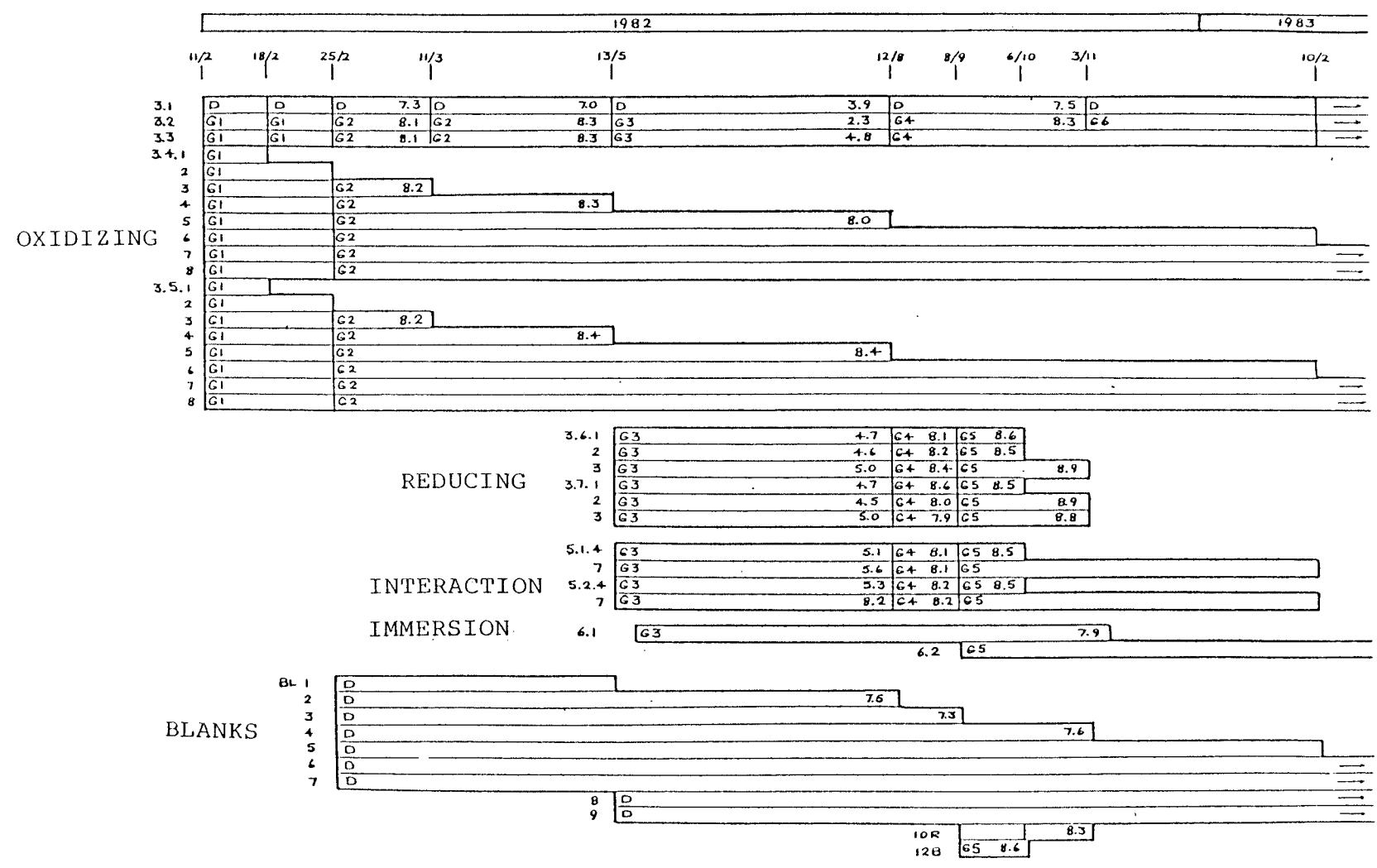


Fig 2. Schematic summary of leach program.

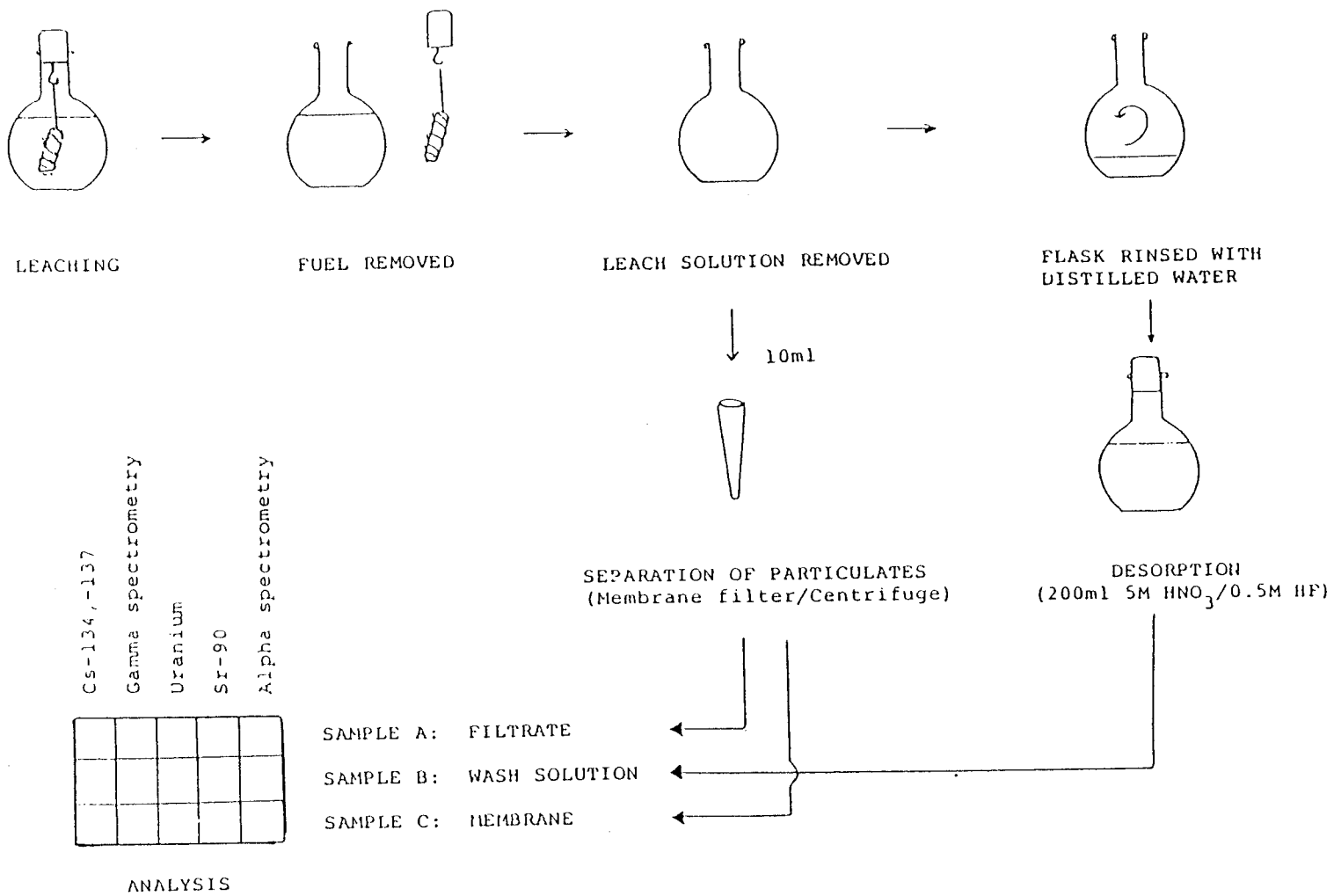


Fig 3. Sketch of procedure for Task 3A leaching.

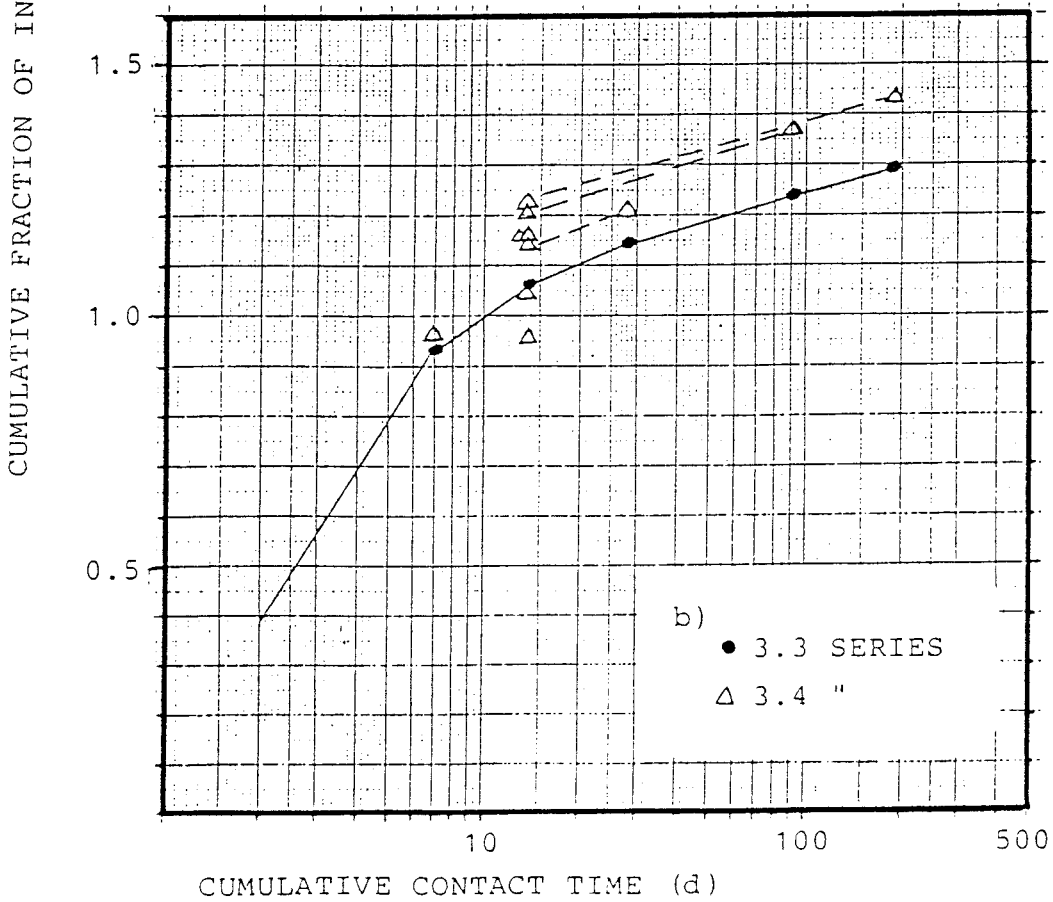
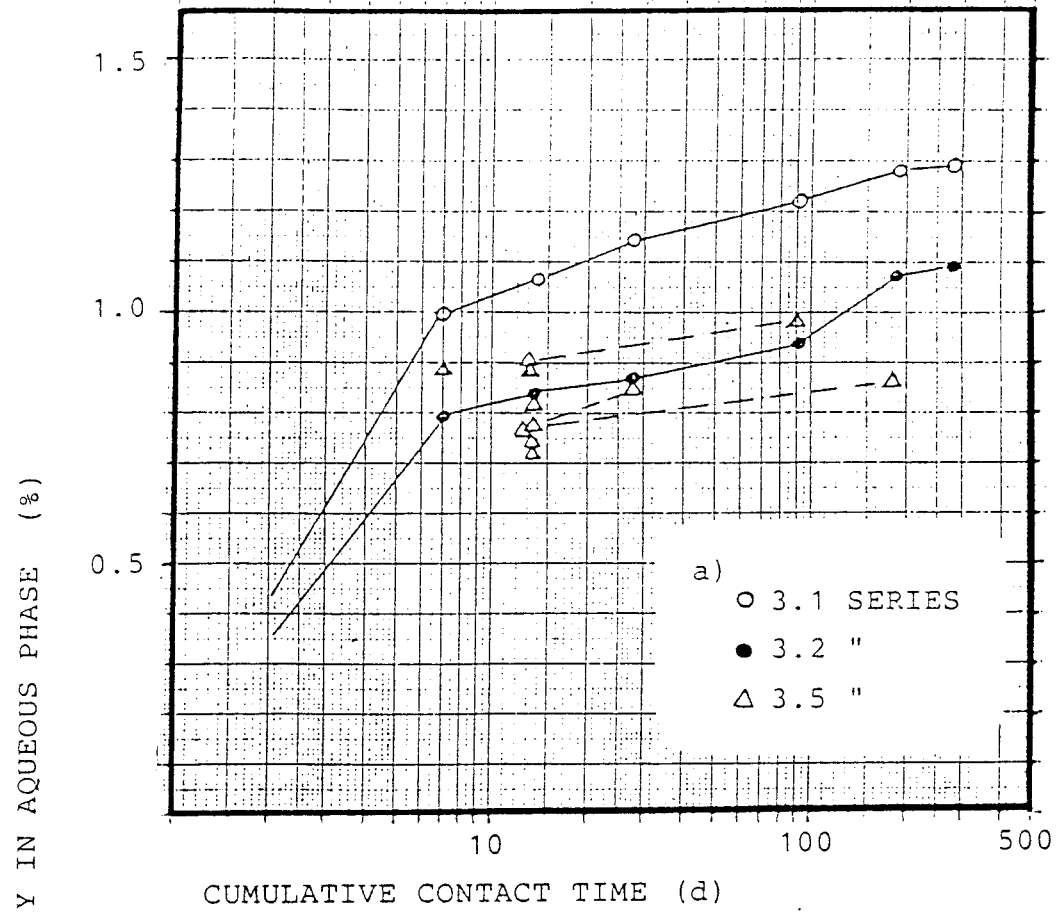


Fig 4. Cs-137 results, showing effect of position of fuel sample.

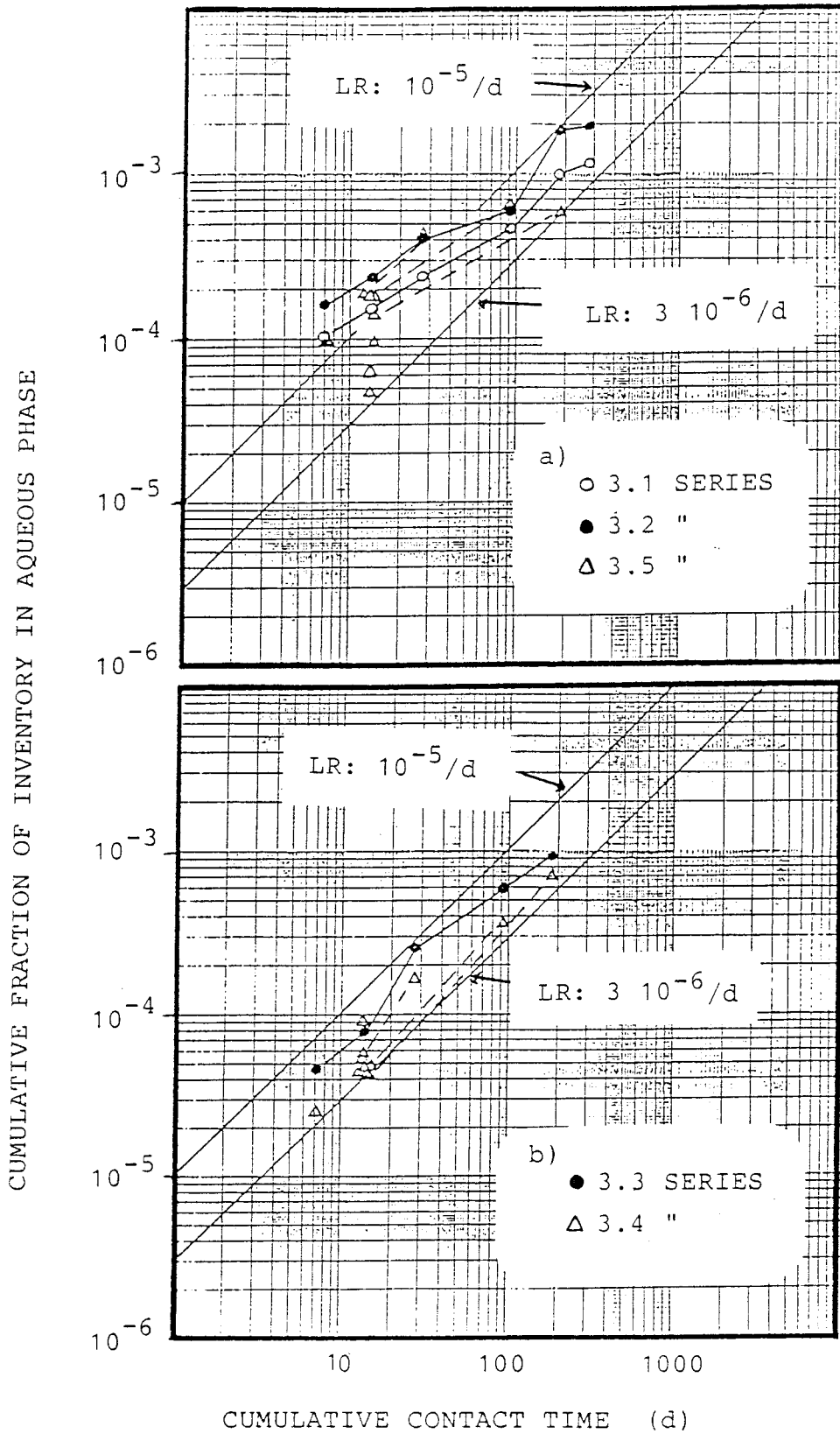


Fig 5. Sr-90 results showing effect of position of fuel sample.

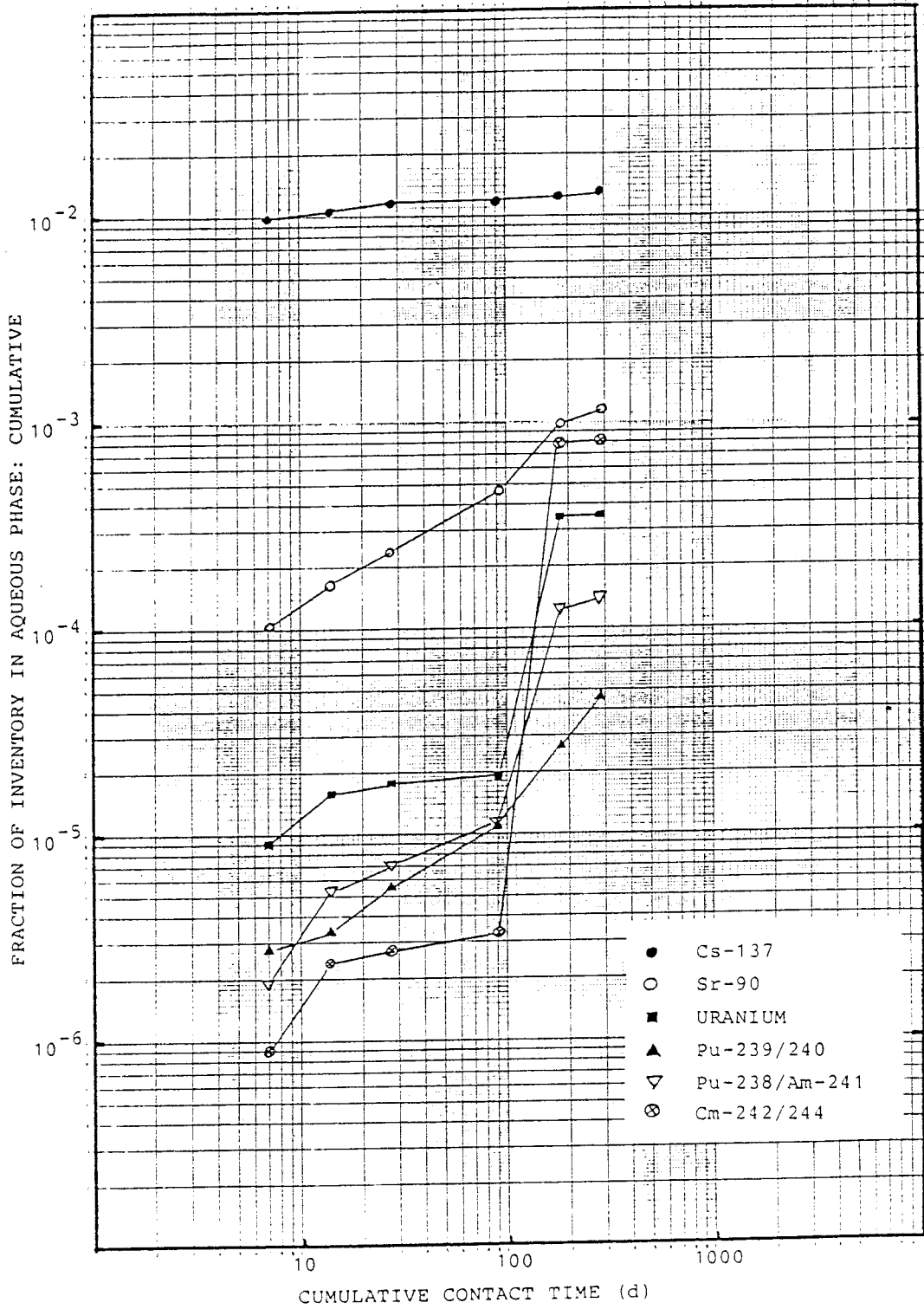


Fig 6. 3.1 SERIES: CUMULATIVE FIAP VALUES

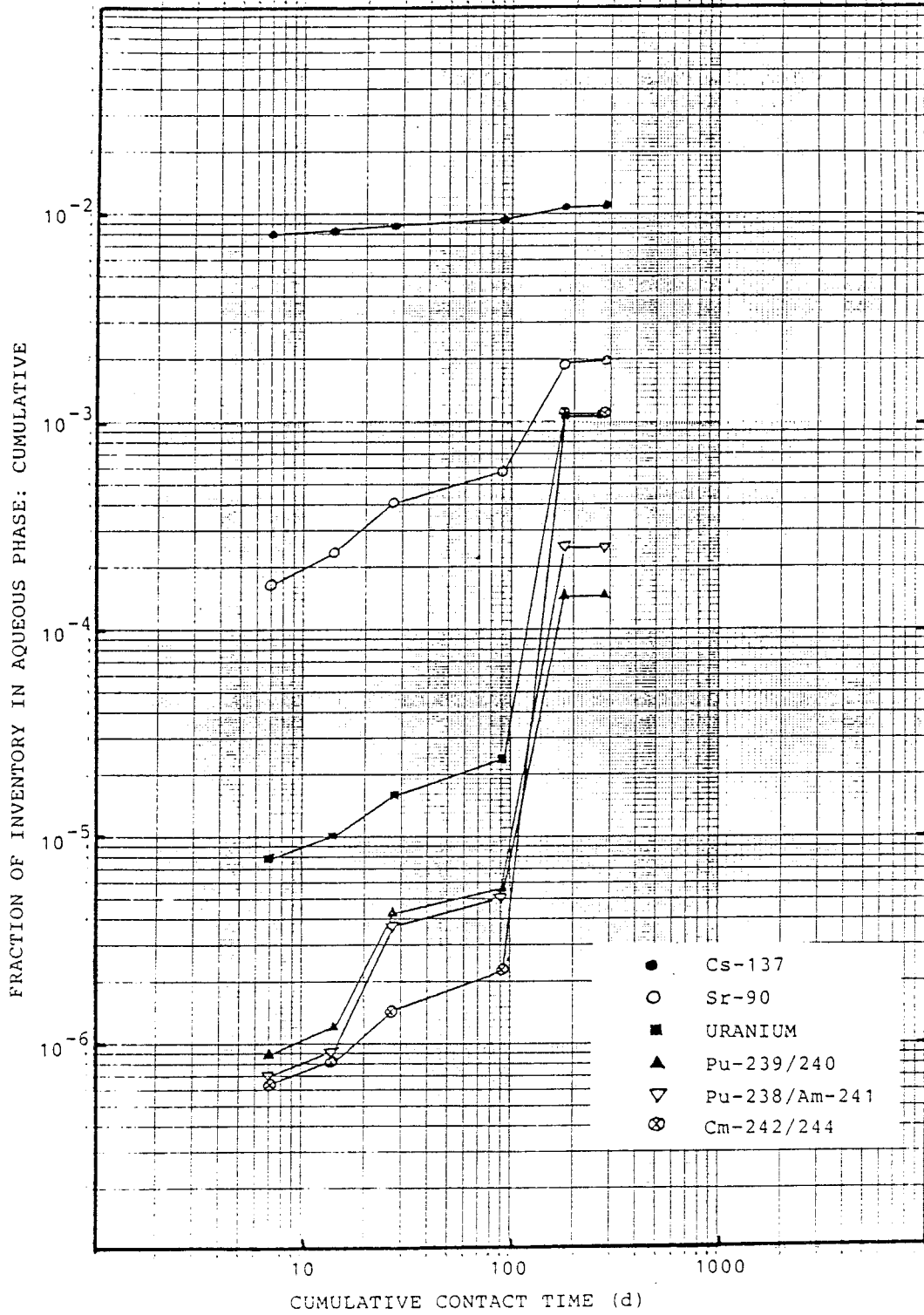


Fig 7. 3.2 SERIES: CUMULATIVE FIAP VALUES



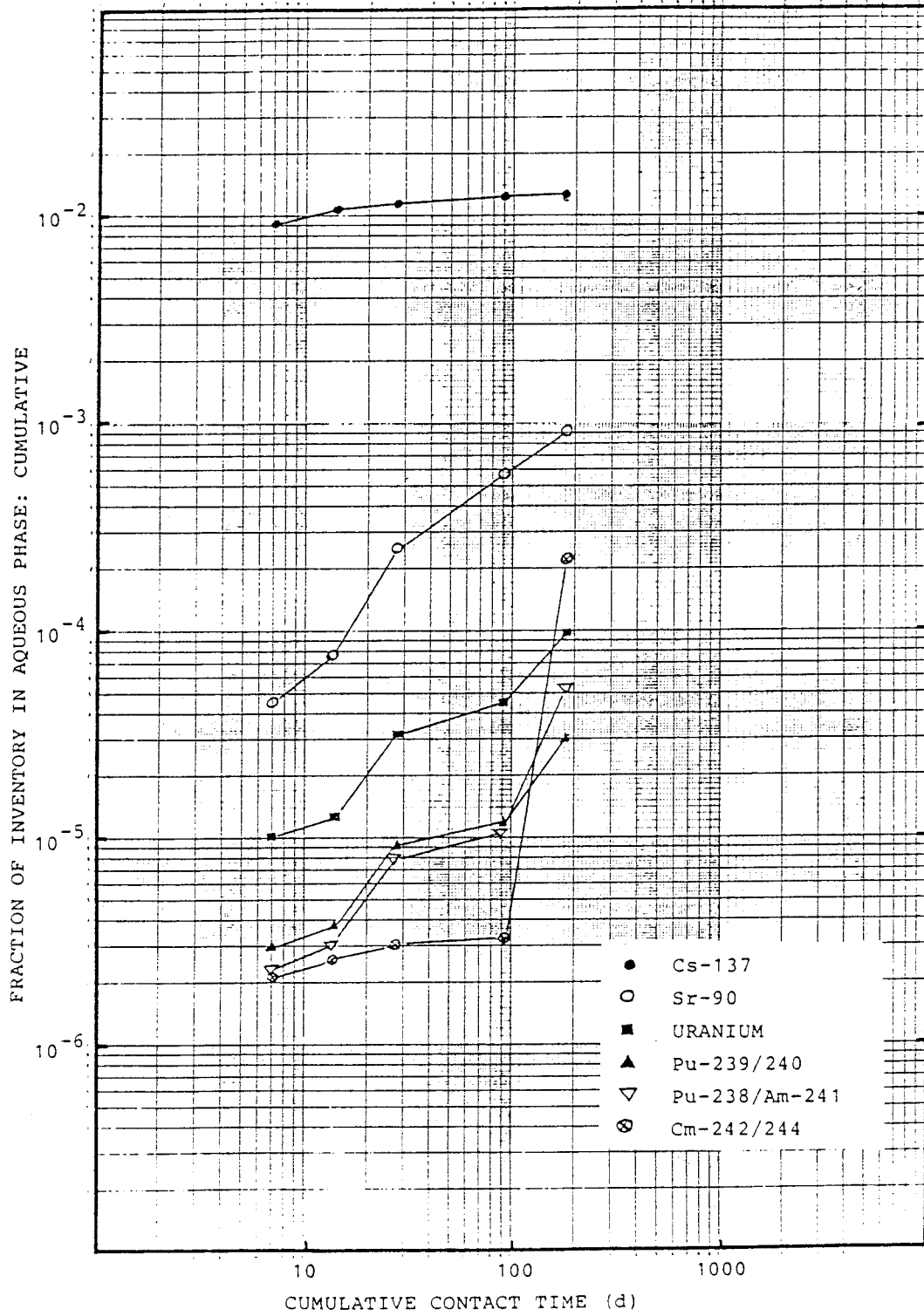


Fig 8. 3.3 SERIES: CUMULATIVE FIAP VALUES

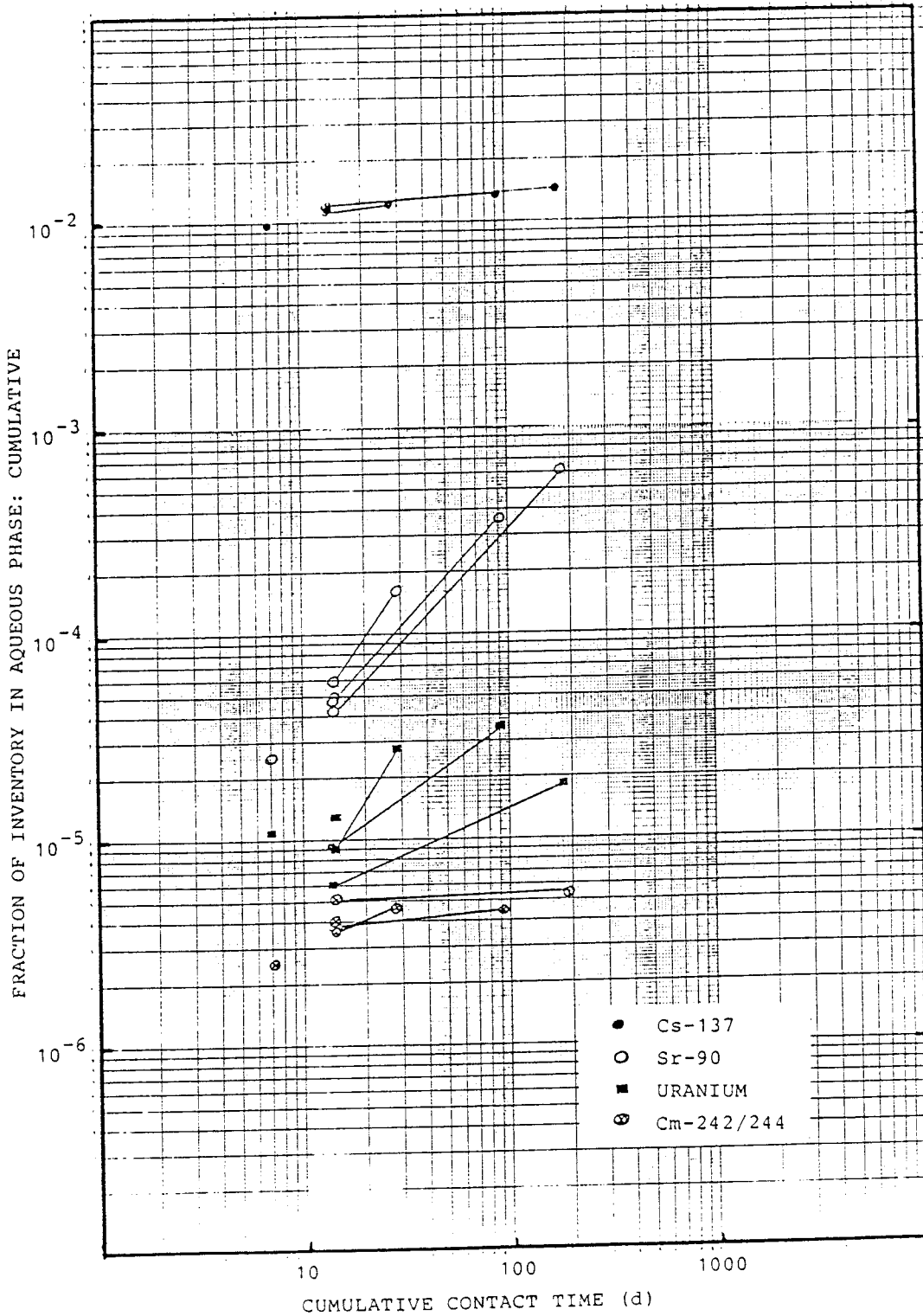


Fig 9. 3.4 SERIES: CUMULATIVE FIAP VALUES

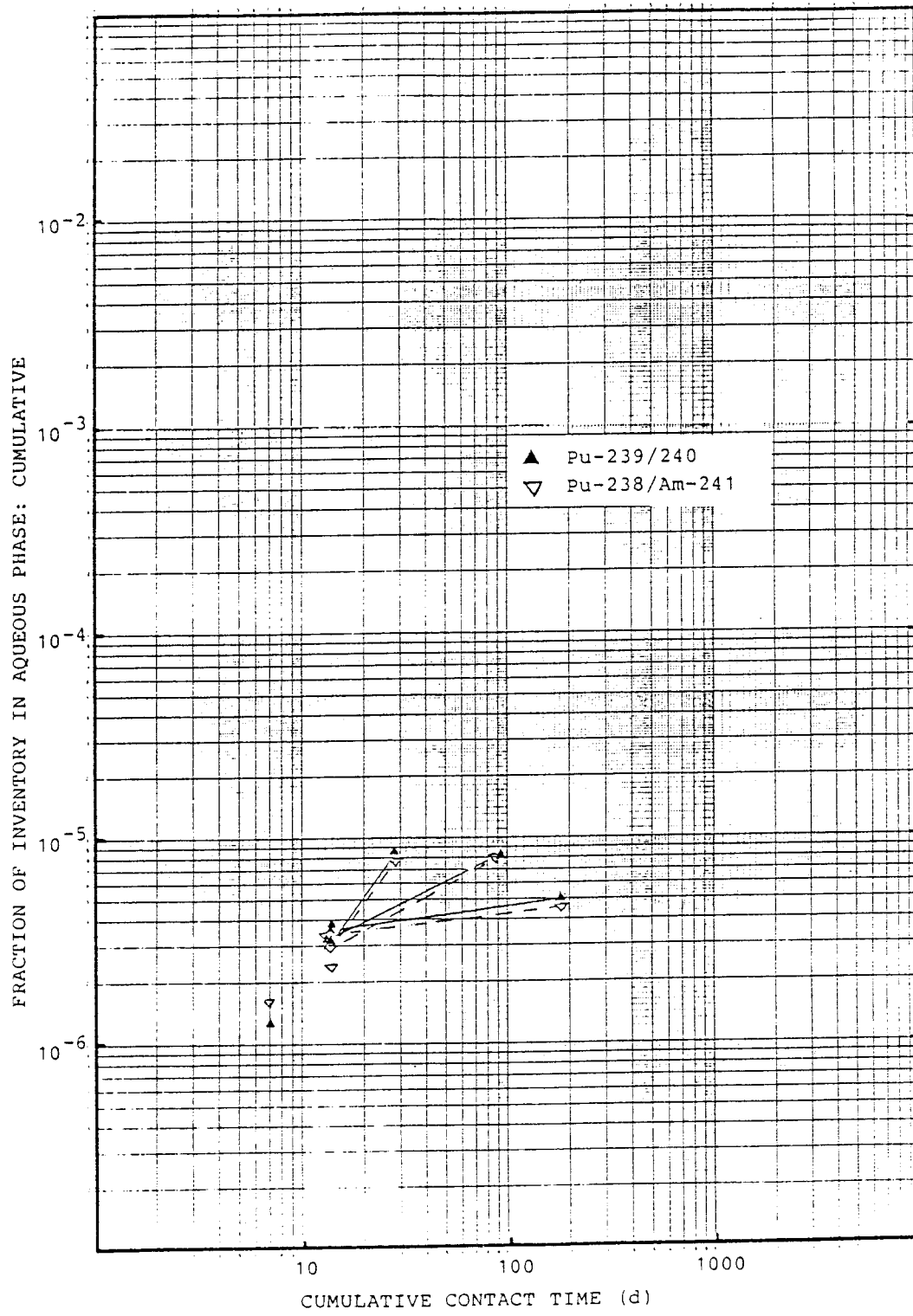


Fig 10. 3.4 SERIES: CUMULATIVE FIAP VALUES  
Pu/Am ISOTOPES

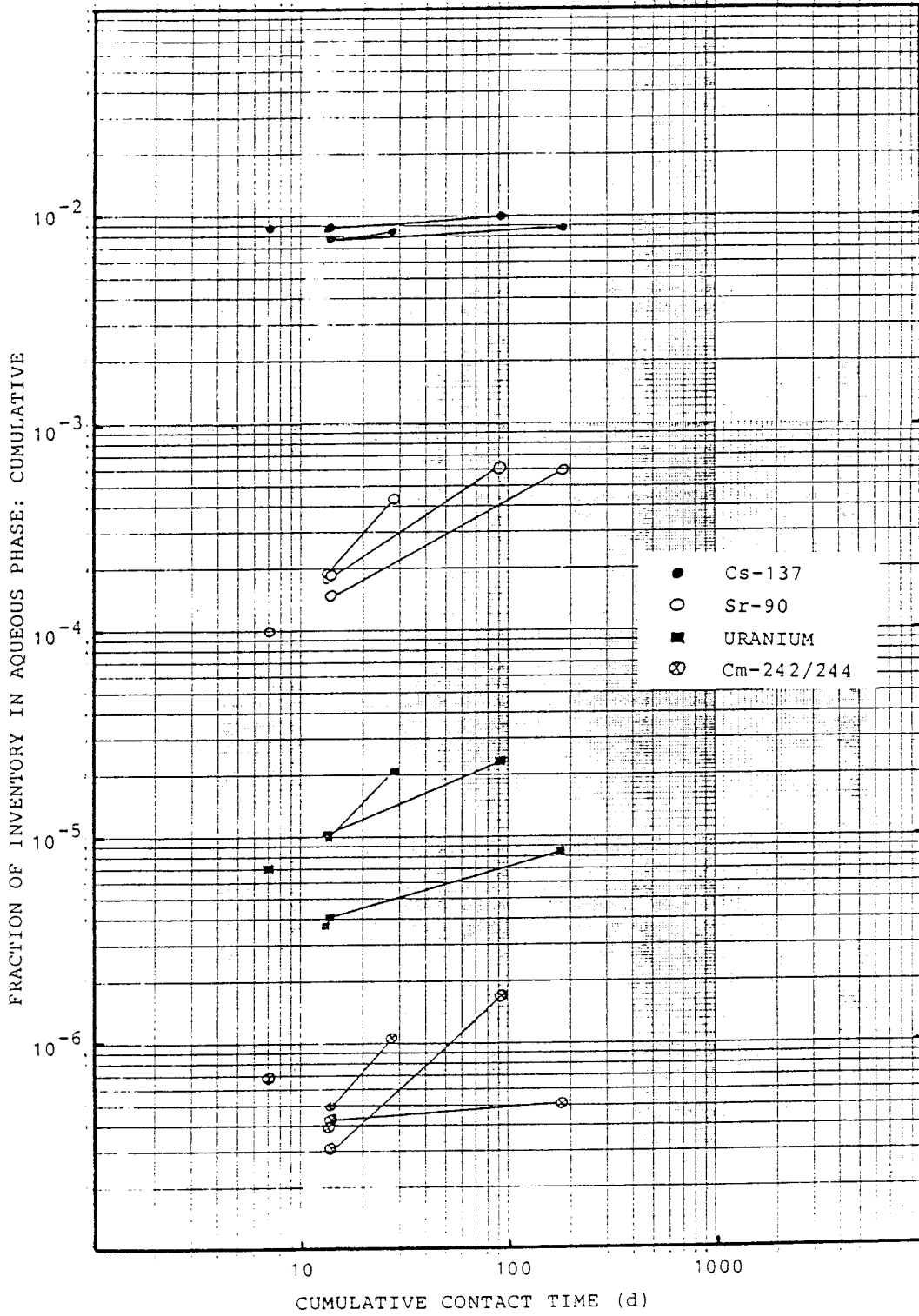


Fig 11. 3.5 SERIES: CUMULATIVE FIAP VALUES

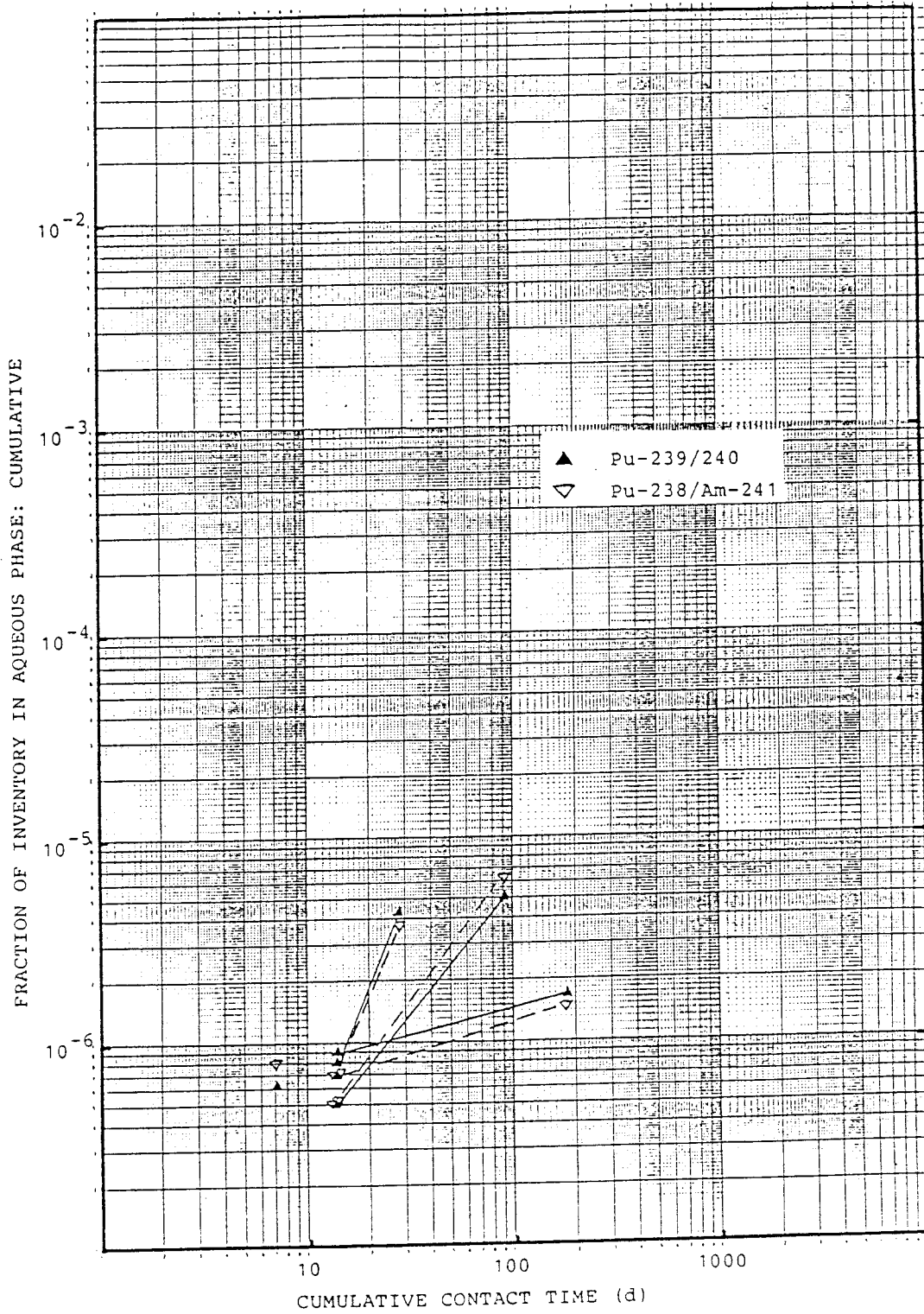


Fig 12. 3.5 SERIES: CUMULATIVE FIAP VALUES  
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