

**SIMFUEL dissolution studies in
granitic groundwater**

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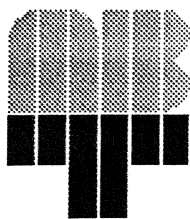
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ABSTRACT

The dissolution behavior of an unirradiated chemical analogue of spent nuclear fuel (SIMFUEL) has been studied in the presence of two different synthetic groundwaters at 25°C and under both oxic and anoxic conditions. The release of U, Mo, Ba, Y and Sr was monitored during static (batch) leaching experiments of long duration (about 250 days). Preliminary results from continuous flow-through reactor experiments are also reported.

The results obtained indicate the usefulness and limitations of SIMFUEL in the study of the kinetics and mechanism of dissolution of the minor components of spent nuclear fuel. Molybdenum, barium and strontium have shown a trend to congruent dissolution with the SIMFUEL matrix after a higher initial fractional release. Yttrium release has been found to be solubility controlled under the experimental conditions.

A clear dependence on the partial pressure of O₂ of the rates of dissolution of uranium has been observed.

INTRODUCTION

In the past ten years an extensive amount of data has been collected within the Swedish nuclear waste management program on the dissolution behavior of UO_2 - nuclear spent fuel. The information obtained from these experiments has been summarized previously [1]. This work was mainly concentrated on ascertaining the mechanisms that regulate the matrix waste dissolution behavior as a function of different groundwater parameters, particularly redox potential and chemical composition. Recently, a model has been proposed for the radiolytically induced oxidative dissolution of spent fuel [2].

It is of great importance to ascertain to what extent and in which way the behavior of the various minor components is related to the oxidative dissolution of the UO_2 spent fuel matrix, particularly in the case of Sr. This element has been proposed as an indicator of matrix dissolution, and arguments have been raised in favor and against this hypothesis [3].

Long term spent fuel dissolution experiments under proper experimental conditions are the most direct approach to any mechanistic study of its behavior. However, the inherent difficulties and limitations associated with the manipulation of spent fuel pellets make them very difficult. Hence, it appears necessary to complement such experiments with other experiences with unirradiated material. This should provide guidance on the mechanistic processes relevant to the dissolution of minor components, as experiments with unirradiated UO_2 do for the behavior of the matrix. In this context, SIMFUEL, a multicomponent UO_2 -based solid solution, could potentially be an appropriate non-active matrix. It should be kept on mind that SIMFUEL is a composite material which attempts to simulate the chemical composition of spent fuel at different levels of burnup, and that, consequently, it constitutes exclusively a chemical analogue of spent fuel. Any further extension of the behavior of SIMFUEL to that of actual spent nuclear fuel should be handled with extreme caution.

The overall objective of the present study is to gain a better understanding of the mechanisms and processes which control the dissolution of the minor components of the spent fuel under realistic repository conditions.

The specific aim of this work is to present and discuss the results so far obtained in the study of the kinetics and thermodynamics of SIMFUEL dissolution at 25°C in granitic groundwaters.

EXPERIMENTAL

Materials

The SIMFUEL pellets were prepared by Chalk River Nuclear Laboratories to simulate fission product inventories nominally representative of spent fuels with burnups of 30 and 50 MWd/kg U. They have an average mass of 8 grams and a measured density of 10.62 g cm^{-3} [4] which results in a geometrical surface area of 4.6 cm^2 . The composition of the SIMFUEL pellets expressed as weight ratios to uranium is listed in Table I.

A careful characterization of the SIMFUEL pellets (for both the 30 and 50 MWd/kg U) has been carried out in Studsvik in the same way as it is done for spent fuel pellets [4]. The results obtained showed that significant differences between typical Swedish spent fuel and SIMFUEL at corresponding burnup levels could be noted regarding the total porosity and the distribution and chemical state of a number of fission product elements. Furthermore, significant grain size differences were found between the two SIMFUEL pellets representative of different burnups, which are not observed in the nuclear fuel. All these factors can in some degree affect the dissolution behavior and they must be taken into account in the discussion of the obtained results.

Table I.- Composition of the SIMFUEL pellets

| | 30 MWd/kg U | 50 MWd/kg U |
|----|--------------------|--------------------|
| Sr | 1.77 E-03 | 2.74 E-03 |
| Y | 4.41 E-04 | 6.46 E-04 |
| Zr | 3.86 E-03 | 5.72 E-03 |
| Mo | 3.16 E-03 | 5.24 E-03 |
| Ru | 2.04 E-03 | 3.80 E-03 |
| Rh | 4.52 E-03 | 6.25 E-03 |
| Pd | 1.25 E-03 | 2.93 E-03 |
| Ba | 2.09 E-03 | 3.68 E-03 |
| La | 1.93 E-03 | 3.08 E-03 |
| Ce | 6.34 E-03 | 8.77 E-03 |
| Nd | 5.97 E-03 | 1.00 E-02 |

Two non-saline synthetic granitic groundwaters were used in these experiments, the so-called Allard groundwater and a more complex one (GW) which attempts to reflect equilibrium with granitic bedrock. The compositions of these groundwaters are given in Table II.

Table II.- Composition of the groundwaters (mol dm⁻³)

| | Allard | Granitic Groundwater |
|-------------------------------|-----------|-------------------------|
| HCO ₃ ⁻ | 1.80 E-03 | 2.75 E-03 |
| Mg | 1.64 E-04 | 9.90 E-05 |
| Si | 2.06 E-04 | 9.95 E-04 |
| Ca | 3.98 E-04 | 6.13 E-04 |
| P | --- | 3.20 E-06 |
| Al | --- | 1.85 E-06 |
| S | --- | 2.10 E-05 |
| pH | 8.2 | 7.5 |

Method

BATCH STUDIES

KTH

The experiments performed at KTH (Royal Institute of Technology, Stockholm) were conducted in two different ways: batch replenishment studies (series B) and batch without replenishment (series C). All of them were made at room temperature, 25 °C.

The first series of tests (B) was conducted using Allard water as leachant and under slightly oxic conditions (the upper 50 ml of the bottle were filled with air). One SIMFUEL specimen was placed into a polypropylene bottle of 150 ml total volume, and 100 ml of leachant were used in each run. The solution was deaerated with N₂ prior to the start of the experiments and the bottles were kept closed during each run. On completion of the specific contact time, the water was removed for analysis and the same solid was put in contact with new solution.

The second series of test was performed under constant O₂ (0.97 atm) and CO₂ (0.01 atm) partial pressures atmosphere. The SIMFUEL pellet was immersed in 1 liter of synthetic granitic groundwater (GW) in a polypropylene bottle. A sequential dissolution procedure was used in these test, in which aliquots (100 ml) were periodically sampled for further analysis.

All of the samples (series B and C) were immediately filtered through 50 nm membrane filters directly into a sample flask. The filtered solutions were analyzed for the various components of both the SIMFUEL and the reference groundwaters.

VII

This series of experiments (Tech. Res. Center of Finland, Espoo) was carried out as follows. The Allard groundwater used for all the experiments was prepared under atmospheric conditions. Then it was deaerated with N₂, transferred into an anaerobic glove box, and allowed to equilibrate for one week prior to the start of the tests. In one series ferrous chloride was added to the water to ensure reducing conditions.

Each SIMFUEL pellet was immersed in 100 ml of the equilibrated leachant in 100 ml Schott Duran flasks. At the completion of contact time, the pellet was removed from the flask and transferred to a new one containing fresh leachant. Aliquots were taken for subsequent analysis and the rest of the solution was filtrated by using ultrafiltration membranes of 3 to 4 nm nominal pore size (Diaflo XM50, Amicon). The filtration was carried out inside the anaerobic box. The pH of the filtrate was measured and the solution was analyzed for the minor components and uranium.

FLOW STUDIES

A continuous flow reactor was developed to contain a SIMFUEL pellet, in which the fresh feed solution was circulated by means of a peristaltic pump and samples of the contacted solution were collected at preset intervals for analysis.

In this kind of reactor the system should evolve to a steady state condition if the flow, the composition of the incoming liquid, the composition of the solid and its surface area remain constant. No formation of secondary phases should take place since the products of the reaction are washed out of the reactor before saturation can be reached. An important characteristic of this system is that the SIMFUEL pellets do not require any manipulation. Once the outer layer of the pellet is dissolved, fresh solid is continuously exposed to the leaching solution.

The same synthetic groundwaters used for the batch experiments (Allard and GW) were used as feed solutions. Two different sets of experiments were conducted as indicated in Table III.

Table III.- Experiments with the continuous flow reactor

| Series | Groundwater | Gas | Pellet (MWd/kgU) |
|--------|-------------|-----------------------------------|------------------|
| F | Allard | N ₂ | 30 |
| L | Allard | N ₂ | 30 |
| N | Allard | N ₂ | 50 |
| G | GW | O ₂ /1%CO ₂ | 30 |
| CB | GW | O ₂ /1%CO ₂ | 50 |
| CM | GW | O ₂ /2%CO ₂ | 50 |

Analytical methods

The analytical methods used for the solution samples from the experiments performed at KTH were as follows. Uranium was analyzed by a Scintrex UA-3 laser fluorescence analyzer, the cations in the synthetic waters were determined by an inductively coupled argon plasma spectrometer (ICP) and the minor components of the SIMFUEL were analyzed by ICP-USN (ultrasound nebulizer). The bicarbonate concentration was determined by standard titration and the pH was measured with a Radiometer pH-meter.

In the experiments performed at VTT, the analysis were done as follows. Uranium and molybdenum were analyzed by Instrumental Neutron Activation Analysis (INAA). Barium and strontium were determined by Atomic Absorption Spectroscopy (AAS), while yttrium was analyzed by ICP. The pH was measured with an Orion Research Expandable Ion Analyzer (model EA 940). The lectures were made against a silver/silver chloride reference electrode.

The oxygen content in the atmosphere within the glove box was measured with a Delta F Corporation Trace Oxygen Analyzer, while its carbon dioxide content was measured with a Fuji Electric Infrared Gas Analyzer.

RESULTS

The concentrations measured as a function of time for the different experiments (both in static and dynamic systems) are presented as Tables in APPENDIX. In the static system, the minor components presented are Mo, Ba, Y, and Sr, since they were readily measured in both laboratories, KTH and VTT, which enables the comparison of the results obtained.

Data treatment

Batch experiments

In order to be able to compare the results obtained in the different experimental procedures, the data were normalized in the following way. In series B (Allard) the concentration of the minor components and uranium was integrated in order to obtain cumulative concentration values.

Then, concentrations of the minor component measured in all experiments (Allard and GW) were normalized to their initial content in the pellet by using the original molar fraction in the solid:

$$\text{Fraction [X]} = [\text{X}] / (\text{weight ratio X to U} * (238.03/\text{molecular weight of X}))$$

The next step consisted of converting these normalized concentrations to total moles of element dissolved. In the so-called Allard series the cumulative values only needed to be multiplied by 0.1, which is the volume (in liters) used in each run. For the GW series, the total number of moles dissolved is the sum of the total amount of each element in the samples plus the remainder left in solution after leach sampling.

In this way, the only parameters that should affect the comparison of the results are either the procedure used and/or the different composition of the test solution and/or the redox conditions.

Flow experiments

In order to compare the data obtained in this system with the results obtained in static experiments, the total amount of element dissolved has to be known. To calculate this value the following procedure was applied. First, the fraction of the concentration measured to the total inventory of the pellet was calculated as in the preceding section. Then, since the flow rate of the feed solution is known, the dissolution rate was calculated as:

$$[X] \text{ (moles/liter)} * Q \text{ (liter/hour)} = \text{moles of X / hour}$$

The sequential amount of element dissolved could then be calculated by multiplying the result obtained by the hours elapsed between two consecutive sample times. Since the concentrations could not be measured continuously, this method assumed that the concentration increases constantly with time. This calculation undoubtedly includes some uncertainty but it was believed to constitute a viable approach. Finally, the sequential concentrations were time-integrated in order to determine the cumulative number of moles dissolved.

DISCUSSION

Batch experiments

In FIGURE 1 the total amount of uranium dissolved in batch experiments is plotted against the total elapsed time.

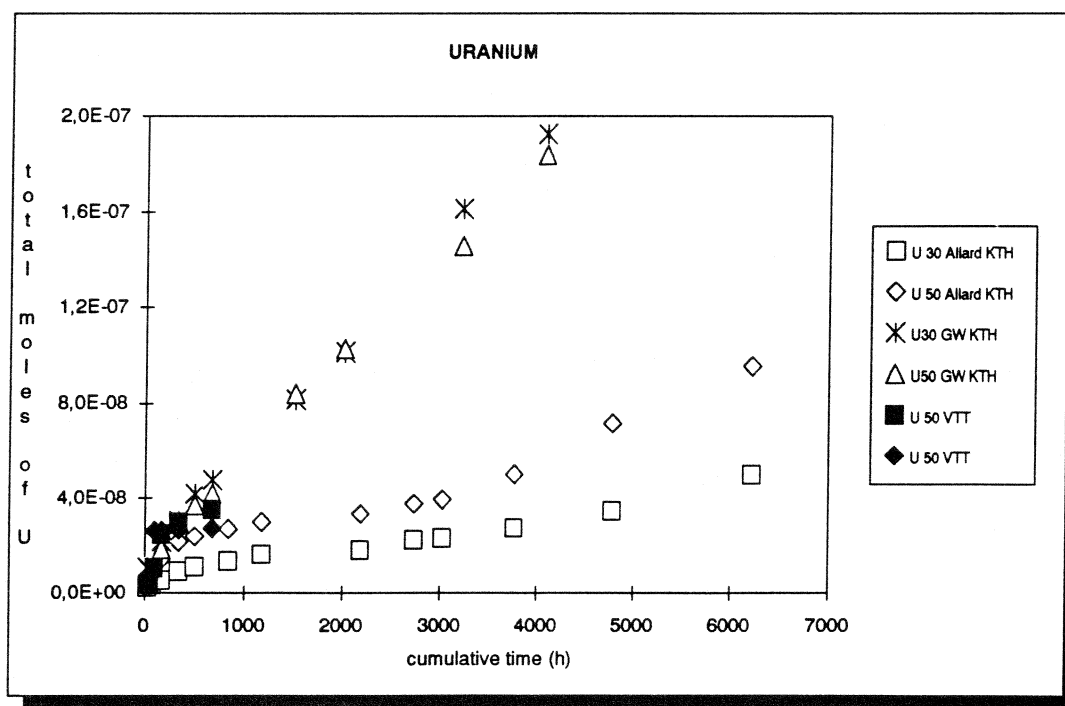


FIGURE 1

We observe significant effects related to the different chemical and redox experimental conditions used, and which can not be attributed to the different experimental procedures followed. It is known that the uranium release can be many times higher in a sequential test than in a static one [2], due to saturation of the leachant in the static case. In our case the results obtained show an opposite behavior. Both

redox conditions and specific surface area of the SIMFUEL pellets vary with respect to the spent fuel leaching experiments. A discussion of the different effects of these parameters on the dissolution rate of SIMFUEL, as reflected in the results tabulated in Table IV, follows.

Table IV.- Rates of uranium release

| Allard | GW | Total surface area | Rate (mol h ⁻¹ m ⁻²) |
|--------|------|---------------------|---|
| U-30 | | 0.12 m ² | 5.06 E-11 |
| U-50 | | " | 1.01 E-10 |
| | U-30 | " | 3.67 E-10 |
| | U-50 | " | 3.54 E-10 |

Redox condition: The redox condition in the Allard experiments was slightly oxidant, with an estimated pO_2 of 0.2 atm, as a result of the contact of the solution with 50 ml of air. In the experiments denominated as GW, pO_2 was 0.97 atm. Unfortunately, the data obtained at VTT cannot be compared in this context, since these experiments have been going for a relatively too short period of time to allow a reliable comparison. From the results shown in Figure 1 and Table IV it is evident that the rate of uranium dissolution from SIMFUEL is approximately 4.5 times larger in the case of the GW experiments. This roughly corresponds to the differences in nominal pO_2 in the two kinds of experiments.

Surface area: The surface area was calculated as the product of the weight of the pellet by the total surface area ($0.015 \text{ m}^2 \text{ g}^{-1}$) as determined by the BET method for a similar material (synthetic UO_2 pellets). Uranium leach rates from spent fuel are largely affected by the specific surface area and reported leach rates range from $1 \text{ E-}05$ to $1 \text{ E-}09 \text{ mol h}^{-1} \text{ m}^{-2}$ [5-9]. The wide spread on dissolution rates for spent fuel under the same experimental conditions is likely to be a result of uncertainties on the estimation of the specific surface area. This produces large errors since the surface of spent fuel is altered randomly after reactor operation. If we calculate a dissolution rate based on the geometrical surface area of the SIMFUEL pellets we obtain a mean dissolution rate of $5.7 \text{ E-}08$ ($\pm 3.7 \text{ E-}08$) $\text{mol h}^{-1} \text{ m}^{-2}$. This value falls in the lower range of reported spent fuel dissolution rates. A better agreement could be obtained if we used the measured BET surface area of actual spent fuel, since the measured porosity of SIMFUEL is less than 1%, while the measured values for LWR fuel are in the range of 5%. Furthermore,

the characterization of the simulated 30 and 50 MWd/kg U SIMFUEL pellets indicates a significant difference in particle size. The 50 MWd/kg U SIMFUEL pellet has a smaller average particle size than the 30 MWd/kg U SIMFUEL pellet [4]. Consequently, the 50 MWd/kg U pellets have a larger surface area and this is reflected in the larger dissolution rates measured in Allard water with $pO_2 = 0.2$ atm. However, this difference is not shown in the experiments carried out at larger partial pressure of oxygen (0.97 atm). A possible explanation would be that at larger oxidation potentials the formation of an altered oxidized layer is so extensive that dominates the mechanism of dissolution of the SIMFUEL, precluding the initial particle size effects.

It is evident from this discussion that both surface area and oxidation potential are key factors controlling the dissolution of the UO_2 matrix in both spent fuel and SIMFUEL.

The determination of the oxidation potential due to radiolysis of spent fuel is difficult, although upper and lower limits can be set to the degree of oxidation caused by radiolysis.

A careful experimental determination of the BET surface area of the exposed spent fuel would eliminate most of the uncertainties on the mechanism and rate of dissolution of spent fuel.

FIGURE 2 reports the molybdenum results expressed as dissolution rate versus total time. Uranium results are included in this FIGURE as a comparison.

The behavior of this minor component is, for all the experiments, in fair agreement, showing no significant difference between either different burnup levels or experimental protocols. The ratio of the normalized release rates of Mo and U seems to converge to unity towards the end of the experiments, following a higher initial dissolution of Mo with respect to U. This convergence can be clearly seen in FIGURE 3, where the fraction of molybdenum dissolved per period of contact time is normalized to the uranium dissolved in the same period and plotted versus the total elapsed experiment time. The value of the ratio tends to unity after approximately 1000 hours (for the experiments carried out under reducing conditions even faster), implying congruent Mo dissolution. The molybdenum dissolution rate after the first 1000 hours was calculated to be $1.6 \text{ E-}10 \text{ mol h}^{-1} \text{ m}^{-2}$ in the experiments using Allard water and $3.5 \text{ E-}9 \text{ mol h}^{-1} \text{ m}^{-2}$ in GW water. These results are comparable to the release rates obtained for uranium, specially with those obtained under slightly oxidant conditions (see Table IV). Furthermore, if the rate of dissolution of molybdenum is calculated only for the

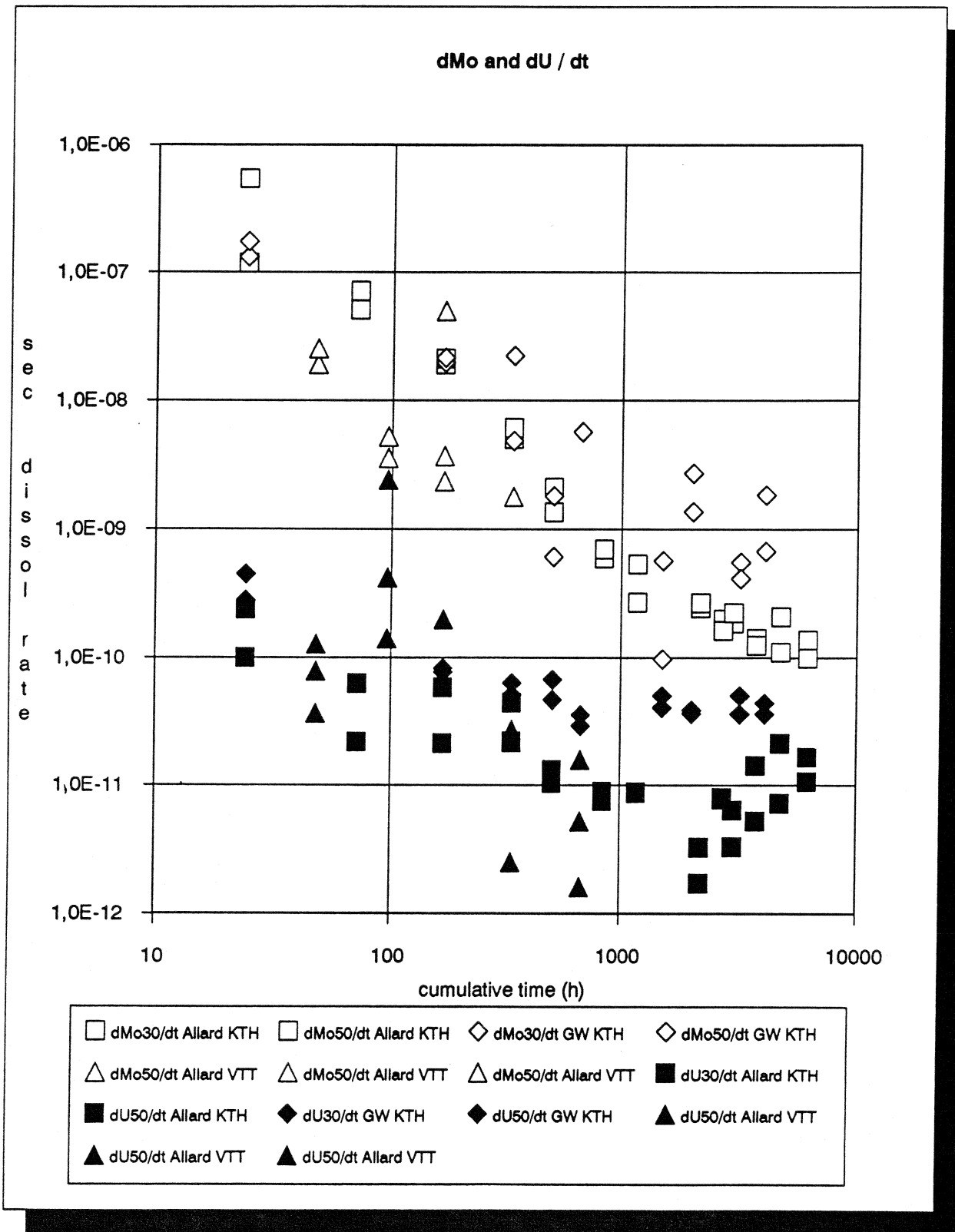


FIGURE 2

first 1000 hours, mean values of $6.2 \text{ E-}09 \text{ mol h}^{-1} \text{ m}^{-2}$ for Allard water and $1.1 \text{ E-}08 \text{ mol h}^{-1} \text{ m}^{-2}$ for GW water are obtained, which are about 50 times higher than those of uranium. This observation gives a semi-quantitative indication of the high initial fraction release cited above.

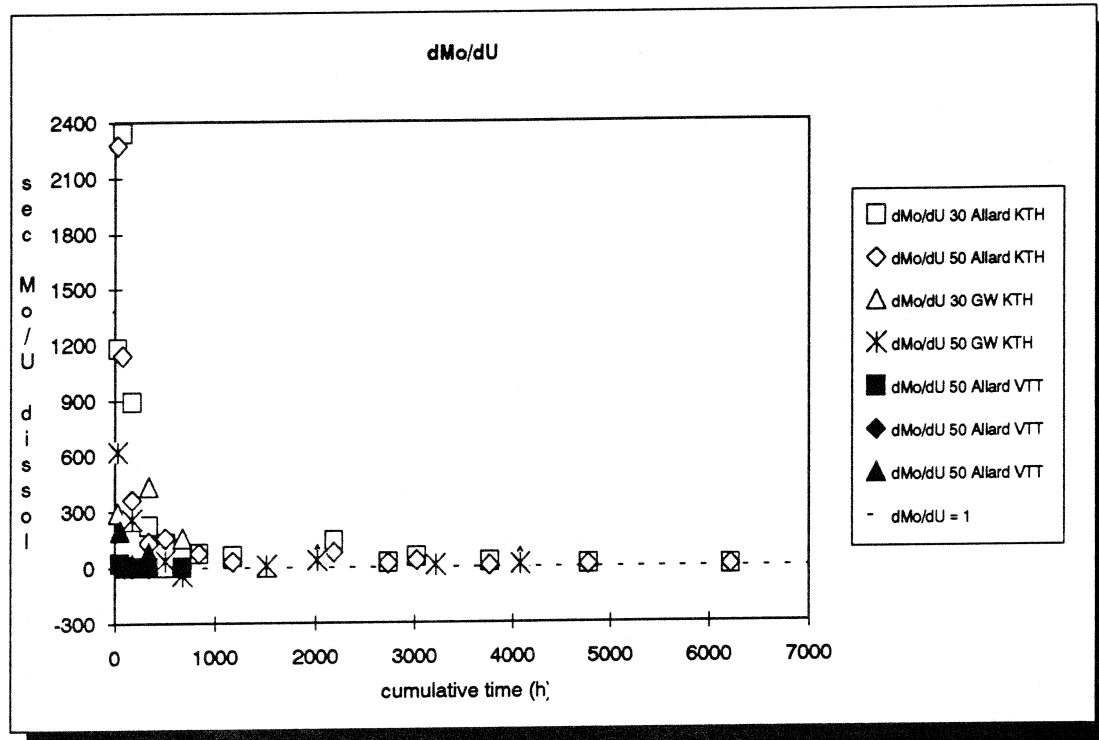


FIGURE 3

According to previous characterizations of SIMFUEL pellets, Mo is present in the form of particles located at the grain boundaries or at grain edges, which appear similar to the metallic fission product particles found in irradiated oxide fuel. Hence, the results obtained with SIMFUEL pellets should be consistent with those obtained with spent LWR fuel leaching experiments. On the other hand, the eventual congruent dissolution of Mo could be an artifact due to subsequent formation of secondary solid phases, either U or Mo phases. In this context it is significant that the characterization of SIMFUEL pellets has shown that a large fraction of Mo is present as MoO_4^{2-} in the solid matrix. If such is the case, the dissolution rate of Mo from the SIMFUEL matrix should be redox sensitive, which would explain the differences in dissolution rates encountered between Allard and GW waters. Neither a characterization of the possible secondary solid phases formed nor an specific study of the effect of the different redox conditions have been done. Future work should focus on these points in order to clarify

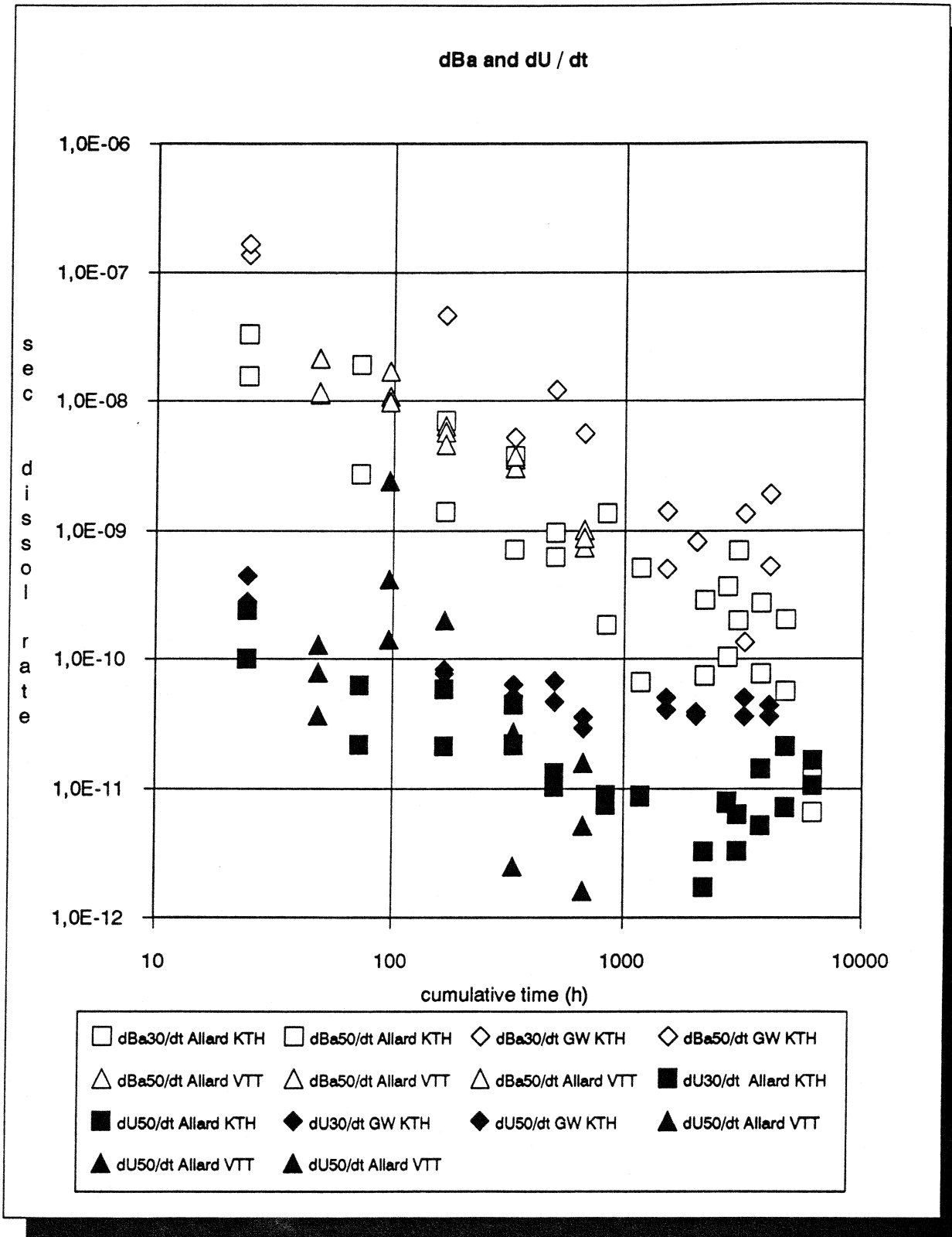


FIGURE 4

achieved after ca. 1000 hours, although in this case the data are somewhat more scattered. In the characterization of SIMFUEL pellets, Ba was found mainly in discrete particles (associated with La, Zr and Sr) of several μm diameter. These phases were not observed at Studsvik in spent LWR fuel.

The significant correlation found between Mo and Ba behavior can indicate either congruent dissolution of both elements with the matrix or the formation of a secondary solid phase controlling both elements. In this case it was postulated [11] the possibility of formation of a barium molybdate phase controlling the release of these elements. Again, future work should concentrate on the localization and characterization of possible secondary phases formed during static leaching.

In FIGURE 6 the yttrium dissolution rate is plotted against total leaching time. For this element only results reported by KTH are presented, since the analytical procedure used in VTT had a detection limit too high for the concentrations found in solution.

The comparison of the release rates found for this minor component and the ones corresponding to uranium shows their analogy. In this case there is no eventual convergence of the dissolution rate with U, as in the cases of Mo and Ba. Instead, Y data seem to follow the same trend as uranium. In FIGURE 7 the fraction of Y released per period of time and normalized to the sequential moles of U released is presented as a function of the total leaching time. The results show considerable scatter and no definite trend can be observed.

A closer look to the yttrium concentrations measured in the different experiments (see Tables in Appendix) shows essentially constant values after different leaching periods. It can be noticed that these values are actually slightly higher in the experiments carried out with Allard water, which has a lower bicarbonate content. These observations lead to speculate that the concentrations measured actually represent saturation of a yttrium solid phase rather than a kinetically controlled release of this minor component. Knowing the bicarbonate and hydroxide concentrations, we compute from literature values for $\text{Y}_2(\text{CO}_3)_3 \cdot 3\text{H}_2\text{O}$ [12] and $\text{Y}(\text{OH})_3$ [13] solubility products a solubility higher than the one observed in these experiments. However, the yttrium concentrations measured in our experiments are very close to the ones expected for Americium and Europium hydroxo and hydroxo carbonate solid phases [14,15 and references therein] (see Table V).

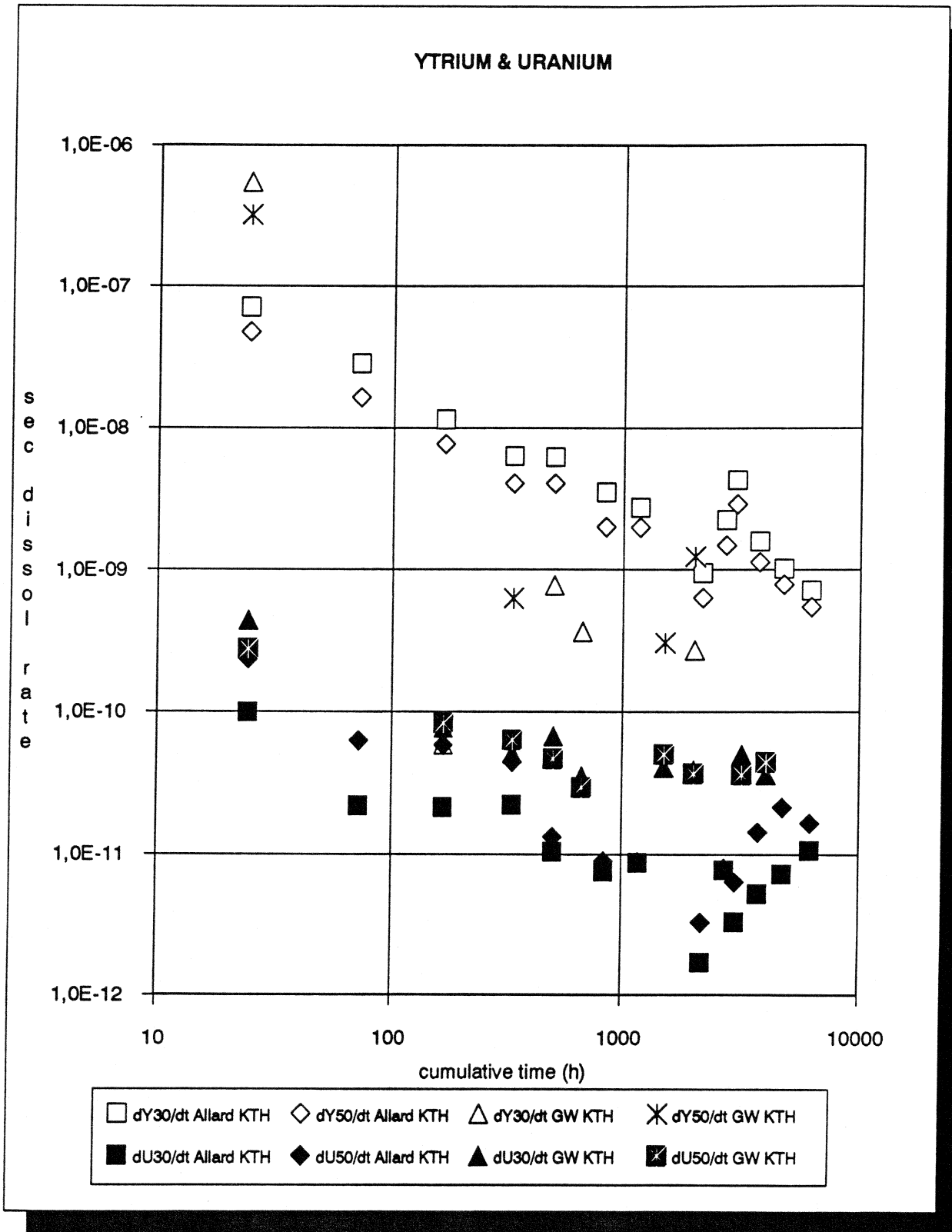


FIGURE 6

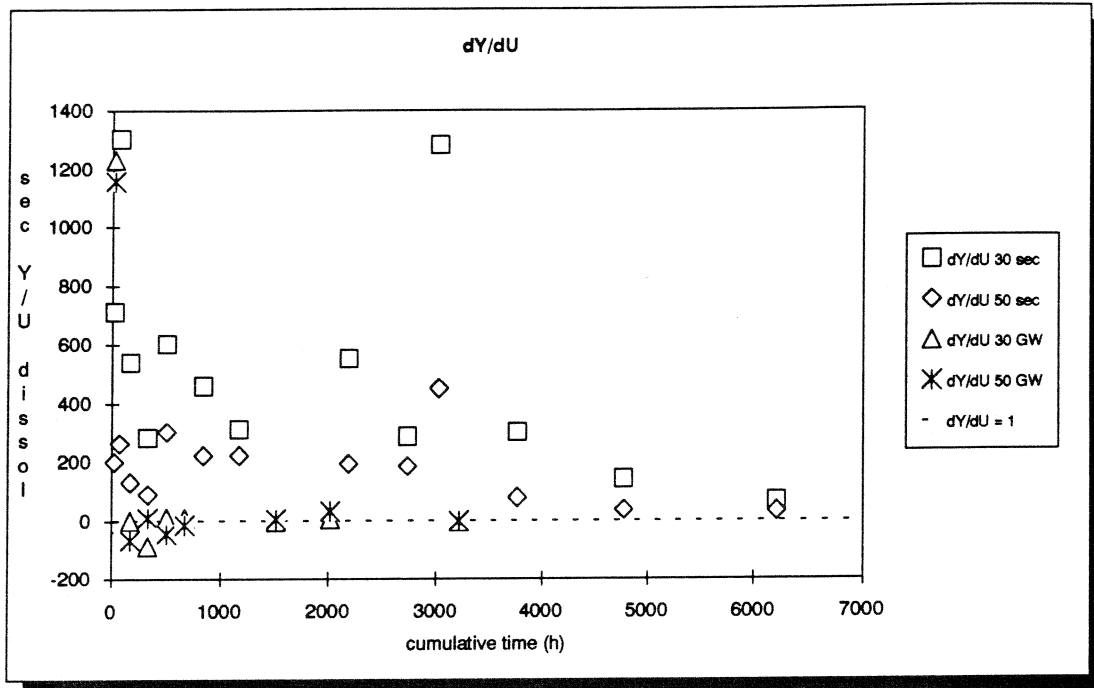


FIGURE 7

Table V.- Yttrium solubilities (mol dm^{-3})

| $\text{Y(OH)}_3(\text{s})$ | $\text{Y}_2(\text{CO}_3)_3 \cdot 3\text{H}_2\text{O}(\text{s})$ | This work | $\text{Am(OH)CO}_3(\text{s}) / \text{Am(OH)}_3(\text{s})$ | $\text{Eu(OH)}_3(\text{s})$ |
|----------------------------|---|------------------------|---|-----------------------------|
| 3.2 E-07 | 1.7 E-07 | 1.4 E-08 (+ 1 E-09) | 1 E-07 - 1 E-09 | 3 E-08 |

In consideration of the fact that the solubilities of hydroxo and hydroxo-carbonate solid phases of trivalent lanthanides and actinides have been significantly lowered by recent investigations while the thermodynamic database of Y has not been reinvestigated for a relatively long time, we suspect that the observed Y concentrations may actually reflect the solubility limits under the experimental conditions, and that further work should be dedicated to reinvestigate and reevaluate the Y thermodynamic database.

Finally, for the static experiments, the data collected for Sr are presented in FIGURE 8 in the same way as for the other minor components.

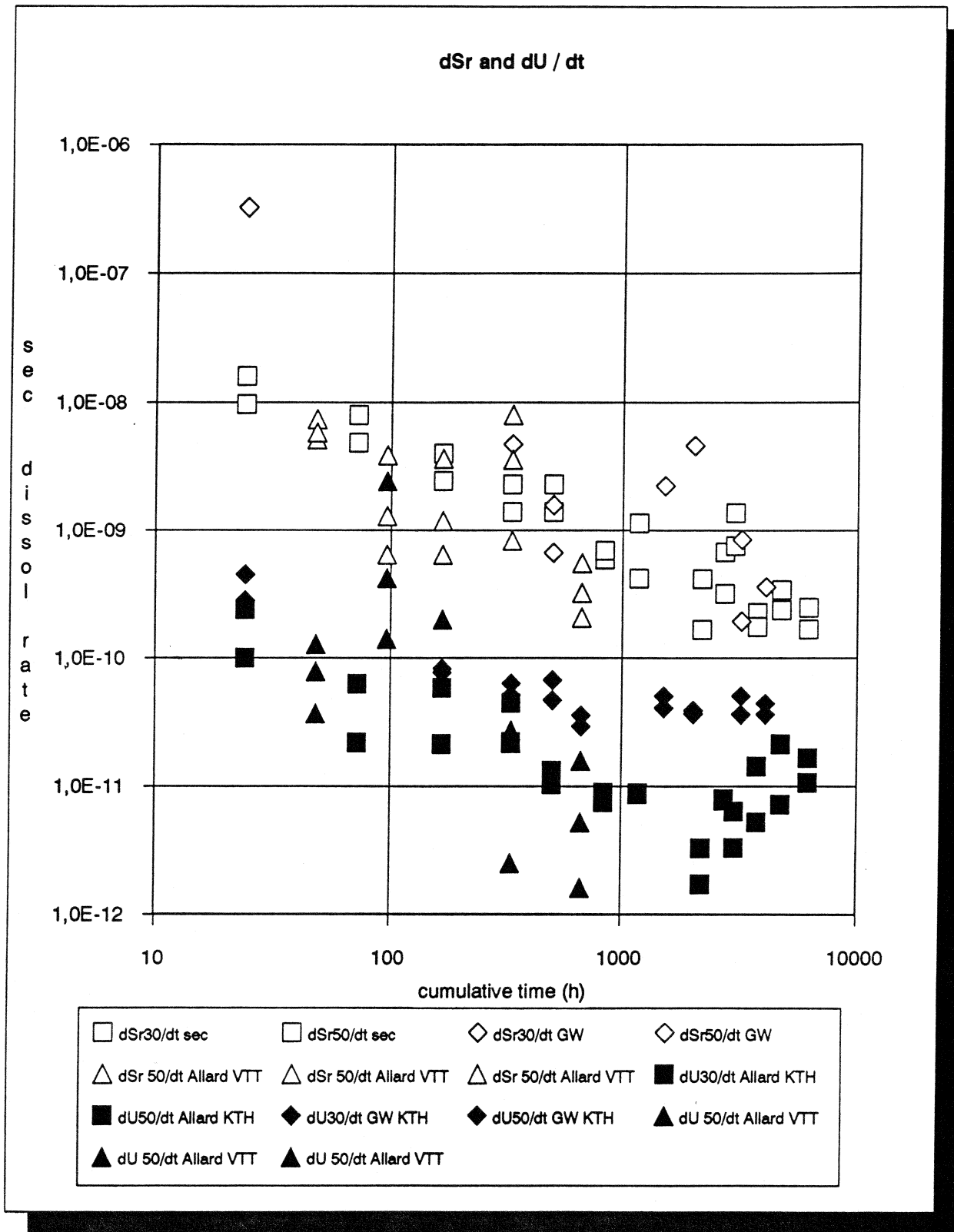


FIGURE 8

The fractional release of strontium follows a behavior parallel to the one obtained for uranium, which is also correlated to the behavior shown by yttrium. On the other hand, when the fractional release per period of time of strontium is normalized to that of uranium (FIGURE 9), after a much higher initial release of Sr respect to U, values tending to one for the Sr/U ratio are obtained very early. The overall Sr release rates were calculated from the data normalized to their initial molar fractions, giving a value of $3.6 \text{ E-}09$ ($\pm 1.1 \text{ E-}09$) $\text{mol h}^{-1} \text{ m}^{-2}$. The relatively close agreement of the overall release rates as well as the convergence towards unity observed in FIGURE 9, point to a matrix congruent dissolution behavior. As indicated in the Introduction section, this element has been proposed by several investigators as a good indicator of fuel matrix dissolution extent, and this is confirmed by the SIMFUEL experiments. On the other hand, it should be noted that Sr is not found in spent LWR fuel in separate particles, as it was found in the characterization of SIMFUEL pellets.

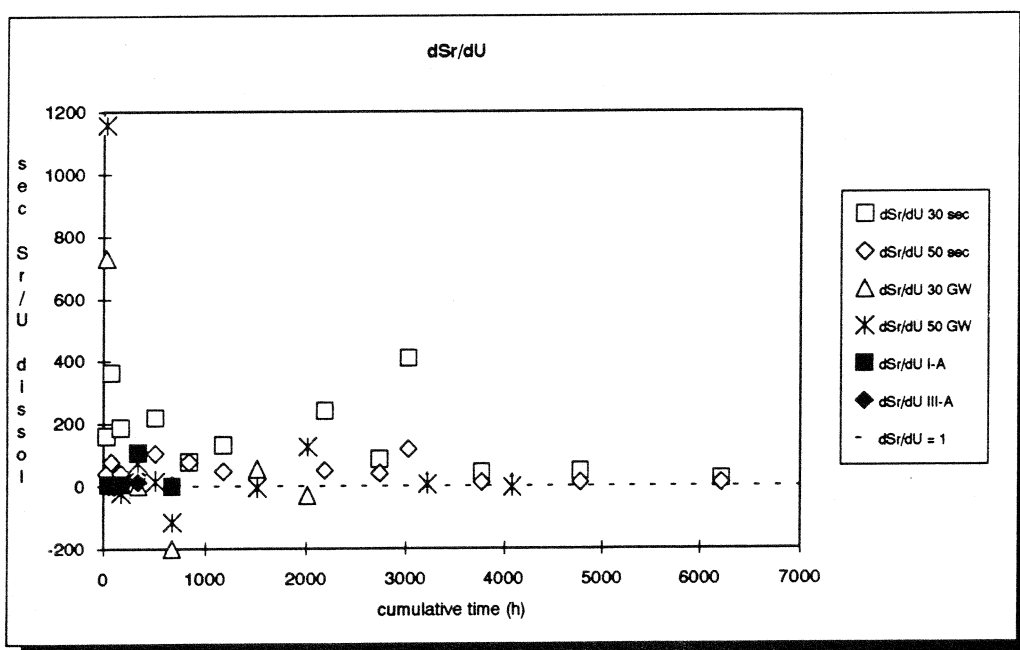


FIGURE 9

Strontium data were plotted in FIGURE 10 as cumulative fraction of inventory in aqueous phase (FIAP) versus contact time. A fairly good agreement can be observed with the results obtained by Forsyth et al. [9] in their leaching experiments with spent nuclear fuel under similar conditions (solid lines in FIGURE 10).

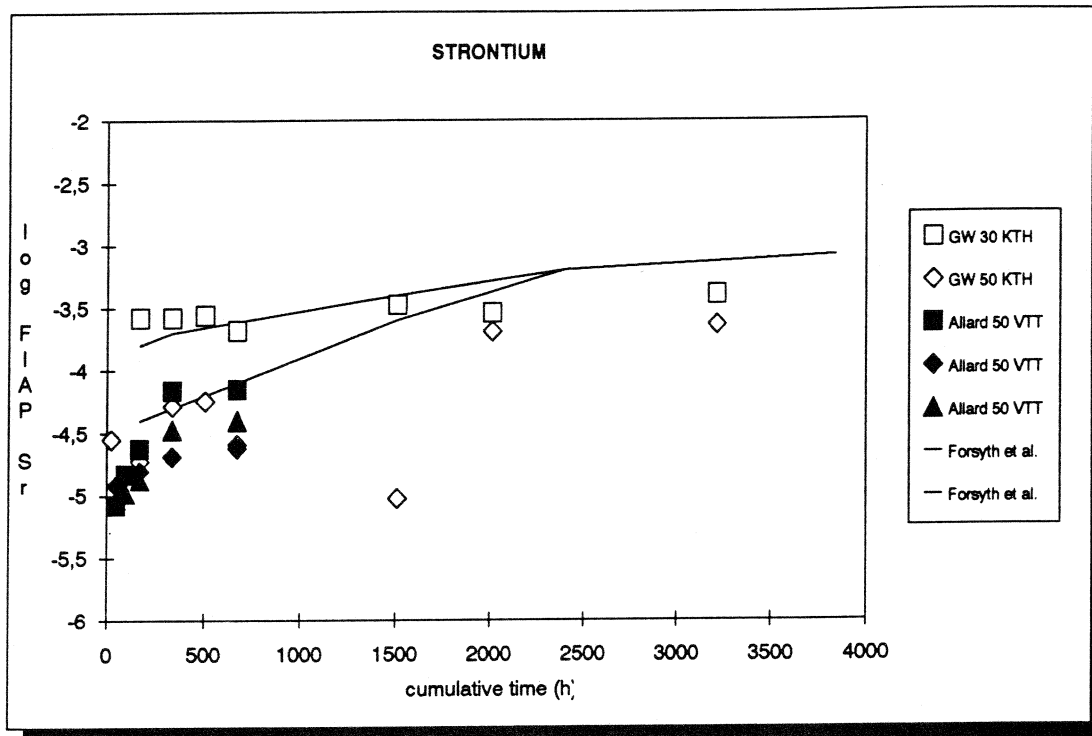


FIGURE 10

Flow experiments

The results obtained in KTH for the two protocols specified in the Experimental section are found in FIGURE 11 for the experiment with Allard water as a leachant and N_2 flux and FIGURE 12 for the one with GW and mixtures of O_2 and CO_2 .

The main conclusions extracted from these data are the following. Regardless of the composition and characteristics of the feed solution, the minor components show similar normalized concentrations in the output solution, whereas uranium appears always at lower concentrations. This can be clearly seen in the values of the corresponding initial release rates calculated, which are presented in Tables VI (Allard water) and VII (GW). The high initial fraction releases of the minor components, compared to uranium, are comparable with the behavior observed at the beginning of the static leaching studies. The relatively short experimental time of the flow reactor studies does not allow to see if congruent dissolution can be eventually obtained in this case. Since secondary solid phases are not expected to be formed in a flow reactor, longer experiences could clarify whether the convergences observed in static leaching experiments correspond to an actual behavior of SIMFUEL dissolution or whether they

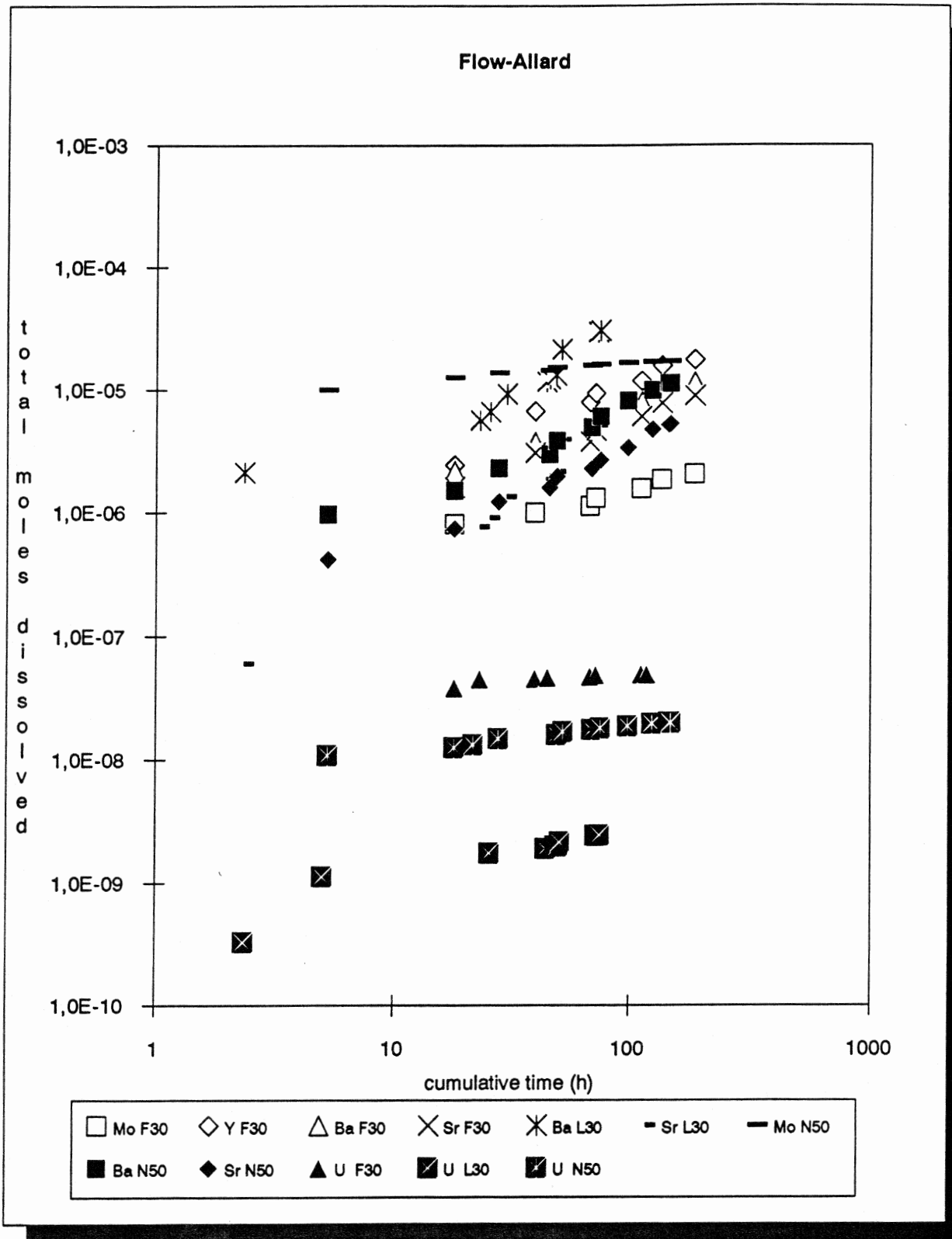


FIGURE 11

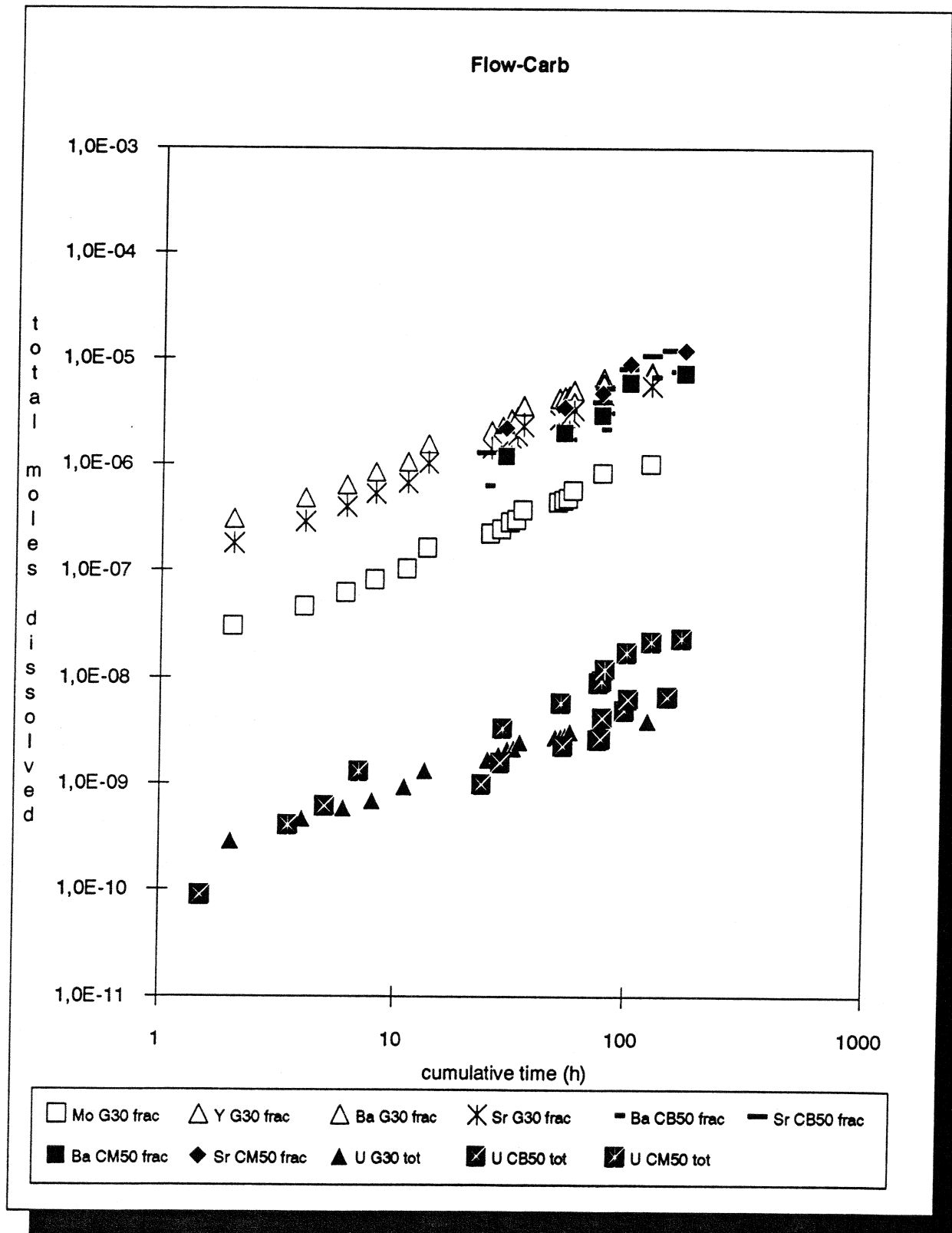


FIGURE 12

are an artifact caused by the formation of secondary solid phases.

Table VI.- Flow experiments with Allard water

| Series | Element | Rate of dissolution (mol h ⁻¹ m ⁻²) |
|--------|---------|---|
| F | Mo | 6.8 E-08 |
| F | Y | 7.5 E-07 |
| F | Ba | 4.9 E-07 |
| F | Sr | 3.8 E-07 |
| L | Ba | 4.1 E-06 |
| L | Sr | 6.4 E-07 |
| N | Mo | 3.3 E-07 |
| N | Ba | 6.4 E-07 |
| N | Sr | 2.9 E-07 |
| F | U | 5.0 E-10 |
| L | U | 2.0 E-10 |
| N | U | 4.8 E-10 |

It can also be seen from these results that the relatively short contact time between the pellet and the solution gives concentrations of yttrium that are lower than those found in the batch experiments. This implies, if the reasoning postulated in the previous section is correct, that this element has not reached saturation in the flow reactor, and that no Y secondary phases are formed. In this case, as it can be seen in FIGURES 11 and 12 and Tables VI and VII, the behavior of yttrium does not differ to the ones observed for the other minor components.

It can be seen in Tables IV, VI and VII that the rate of uranium release is similar in both static and dynamic experiments.

Table VII.- Flow experiments with GW water

| Series | Element | Rate of dissolution (mol h ⁻¹ m ⁻²) |
|--------|---------|---|
| G | Mo | 7.2 E-08 |
| G | Y | 6.8 E-07 |
| G | Ba | 5.2 E-07 |
| G | Sr | 4.0 E-07 |
| CB | Ba | 5.0 E-07 |
| CB | Sr | 7.5 E-07 |
| CM | Ba | 3.7 E-07 |
| CM | Sr | 5.9 E-07 |
| G | U | 2.8 E-10 |
| CB | U | 3.9 E-10 |
| CM | U | 1.3 E-09 |

Finally, it should be pointed out that the reactor used has not shown in some cases an ideal behavior, probably due to its relatively high volume, necessary to contain the SIMFUEL pellet. This non-ideal behavior was clearly demonstrated in some cases when the flow rate of the feed solution was changed. If steady state conditions are reached, rates of dissolution can be directly derived multiplying the flow rate by the element concentration in the output solution. If flow rate is changed, and the reactor behaves ideally, this product must remain constant, which was not observed in the present work in some cases. As a consequence, it is not possible to calculate the rate of dissolution at each sampling. Anyhow, it has also been demonstrated above that the calculation of the overall dissolution rates gave values consistent with static experiment determinations.

The results obtained with this experimental setup have shown that its use could clarify some aspects related to SIMFUEL leaching. Future work orientated to the use of a flow reactor is encouraged, although its design should be improved in order to obtain the maximum reliable information.

CONCLUSIONS

The investigation of SIMFUEL dissolution behavior reported here has shown the potential usefulness as well as the limitations of this chemical analogue in the study of the mechanism of dissolution of spent nuclear fuel.

For molybdenum, barium and strontium, batch dissolution studies have shown a high initial fraction release, followed, after approx. 1000 hours, by a trend to congruent dissolution with the SIMFUEL matrix (UO_2).

Yttrium release is solubility controlled in the batch experiments under the experimental conditions. The concentrations found in solution are not consistent with the yttrium thermodynamic database, but correlate well with the solubility of Am and Eu hydroxo and hydroxo carbonate solid phases under similar experimental conditions.

The determination of the dissolution rate of both SIMFUEL and spent fuel matrices (UO_2) needs accurate surface area determinations (ie. BET measurements) in order to correlate the values obtained using differently altered solid phases. Such determinations would eliminate most of the uncertainties on the mechanisms and rates of dissolution of spent fuel due to the different porosities, cracks and fissures formed during nuclear reactor operation, which increase the total surface area accessible to both leachant and oxidant species.

A clear influence of the partial pressure of O_2 on the rate of uranium dissolution was observed. Although the determination of the oxidation potential due to radiolysis is difficult, the results obtained from SIMFUEL indicate upper and lower limits of dissolution rate that can be expected under different redox conditions.

Flow experiments have shown that reliable results can be obtained using this experimental set-up. A quantification of the initial fraction release rate of the SIMFUEL components has been obtained. Furthermore, since secondary solid phases are not formed in a flow reactor, the comparison of the results obtained in the batch experiments with long term flow experiments may allow to distinguish the eventual role of such solid phases in the mechanism of dissolution of SIMFUEL.

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APPENDIX**TABLE A.1.- KTH: Concentrations measured in Allard water (ppb)**

| t (h) | U ₃₀ | Mo ₃₀ | Y ₃₀ | Ba ₃₀ | Sr ₃₀ | U ₅₀ | Mo ₅₀ | Y ₅₀ | Ba ₅₀ | Sr ₅₀ |
|-------|-----------------|------------------|-----------------|------------------|------------------|-----------------|------------------|-----------------|------------------|------------------|
| 24 | 5,69 | 21,18 | 1,82 | 3,88 | 1,60 | 13,57 | 161,9 | 1,73 | 3,24 | 1,50 |
| 72 | 2,50 | 18,52 | 1,46 | 4,46 | 1,60 | 7,14 | 42,71 | 1,20 | 1,14 | 1,50 |
| 168 | 4,86 | 13,73 | 1,18 | 3,30 | 1,60 | 13,28 | 25,22 | 1,12 | 1,18 | 1,50 |
| 336 | 8,83 | 6,29 | 1,13 | 3,09 | 1,60 | 17,73 | 12,76 | 1,02 | 1,05 | 1,50 |
| 504 | 4,14 | 1,70 | 1,12 | 0,80 | 1,60 | 5,28 | 4,39 | 1,02 | 0,92 | 1,50 |
| 840 | 6,05 | 1,49 | 1,25 | 2,27 | 0,83 | 7,12 | 2,90 | 1,01 | 0,54 | 1,50 |
| 1176 | 6,95 | 1,35 | 0,97 | 0,85 | 1,60 | 7,09 | 1,13 | 1,01 | 0,19 | 0,90 |
| 2184 | 4,12 | 1,86 | 1,02 | 1,43 | 1,75 | 7,90 | 3,37 | 0,97 | 0,66 | 1,08 |
| 2736 | 10,16 | 0,83 | 1,31 | 1,00 | 1,56 | 10,45 | 1,12 | 1,24 | 0,50 | 1,13 |
| 3024 | 2,28 | 0,40 | 1,31 | 1,00 | 1,64 | 4,38 | 0,80 | 1,25 | 0,50 | 1,40 |
| 3768 | 9,31 | 0,79 | 1,26 | 1,00 | 0,70 | 25,23 | 1,16 | 1,27 | 0,50 | 0,84 |
| 4778 | 17,35 | 0,84 | 1,10 | 1,00 | 1,44 | 51,17 | 2,61 | 1,20 | 0,50 | 1,54 |
| 6218 | 36,41 | 1,07 | 1,10 | 0,09 | 1,50 | 56,64 | 2,47 | 1,20 | 0,08 | 1,56 |

TABLE A.2.- KTH: Concentrations measured in GW (ppb)

| t (h) | U ₃₀ | Mo ₃₀ | Y ₃₀ | Ba ₃₀ | Sr ₃₀ | U ₅₀ | Mo ₅₀ | Y ₅₀ | Ba ₅₀ | Sr ₅₀ |
|-------|-----------------|------------------|-----------------|------------------|------------------|-----------------|------------------|-----------------|------------------|------------------|
| 24 | 2,54 | 2,39 | 1,41 | 1,63 | 3,30 | 1,59 | 5,19 | 1,17 | 3,54 | 0,54 |
| 168 | 5,46 | 4,76 | 1,41 | 5,23 | 3,26 | 4,74 | 9,49 | 1,04 | 3,15 | 0,36 |
| 336 | 8,02 | 8,28 | 1,31 | 5,02 | 3,27 | 7,91 | 10,74 | 1,06 | 4,10 | 1,00 |
| 504 | 11,85 | 8,39 | 1,33 | 6,44 | 3,43 | 10,58 | 11,29 | 0,98 | 3,90 | 1,10 |
| 672 | 14,22 | 9,58 | 1,34 | 7,21 | 2,59 | 12,52 | 10,85 | 0,97 | 3,48 | 0,49 |
| 1512 | 30,38 | 9,70 | 1,34 | 8,37 | 4,16 | 32,70 | 12,03 | 1,04 | 4,22 | 0,18 |
| 2016 | 42,20 | 12,27 | 1,37 | 8,89 | 3,57 | 43,49 | 14,18 | 1,28 | 3,16 | 3,92 |
| 3216 | 89,94 | 13,92 | 1,36 | 11,56 | 5,00 | 77,93 | 16,24 | 1,27 | 3,64 | 4,42 |
| 4080 | 126,8 | 19,88 | 1,36 | 15,64 | 5,64 | 123,1 | 19,84 | 1,27 | 5,64 | 3,97 |

**TABLE A.3.- VTT: Concentrations measured in
Allard water (ppb)**

| t (h) | Mo ₅₀ | Mo ₅₀ | Mo ₅₀ | Ba ₅₀ | Ba ₅₀ | Ba ₅₀ |
|-------|------------------|------------------|------------------|------------------|------------------|------------------|
| 48 | 12,52 | 15,20 | 43,98 | 4,70 | 8,90 | 4,90 |
| 96 | 4,16 | 3,12 | 2,09 | 4,50 | 4,10 | 7,10 |
| 168 | 2,07 | 2,11 | 0,42 | 4,00 | 3,60 | 2,90 |
| 336 | 0,40 | | 0,81 | 5,20 | 4,50 | 5,50 |
| 672 | 2,44 | | 0,41 | 2,20 | 3,00 | 2,60 |

TABLE A.3.- (cont.)

| t (h) | Sr ₅₀ | Sr ₅₀ | Sr ₅₀ | U ₅₀ | U ₅₀ | U ₅₀ |
|-------|------------------|------------------|------------------|-----------------|-----------------|-----------------|
| 48 | 1,6 | 2,3 | 1,8 | 9,03 | 14,60 | 4,20 |
| 96 | 1,2 | 0,4 | 0,2 | 16,20 | 47,79 | 276 |
| 168 | 1,7 | 0,3 | 0,55 | 34,20 | | 15,40 |
| 336 | 8,7 | 0,9 | 3,9 | 10,90 | 1,01 | 8,70 |
| 672 | 0,45 | 0,7 | 1,2 | 12,70 | 1,29 | 4,20 |

TABLE A.4.- Flow experiments with Allard water (ppb)

| t (h) | Mo ^F | Y ^F | Ba ^F | Sr ^F | Ba ^L | Sr ^L | Mo ^N | Ba ^N | Sr ^N | U ^F | U ^L | U ^N |
|-------|-----------------|----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|----------------|----------------|----------------|
| 2,3 | | | | | 7,8 | 0,2 | | | | | 1,6 | 16,9 |
| 5,3 | | | | | | | 81,8 | 5,4 | 1,8 | | 1,2 | 3,7 |
| 18 | 1,8 | 0,7 | 3,1 | 2,0 | | | 21,9 | 3,1 | 1,4 | | | 2,5 |
| 23 | | | | | 9,6 | 1,6 | | | | 37,2 | | |
| 25,5 | | | | | 10,4 | 1,3 | | | | | | |
| 27,8 | | | | | | | 8,0 | 3,7 | 1,8 | 12,5 | | 2,04 |
| 30 | | | | | 10,9 | 1,5 | | | | | 0,6 | |
| 39,5 | 0,5 | 1,5 | 2,6 | 2,1 | | | | | | | | |
| 44 | | | | | 11,8 | 2,1 | | | | 0,9 | | |
| 45,7 | | | | | 11,8 | 2,0 | 6,2 | 4,2 | 1,7 | | 0,2 | |
| 48,5 | | | | | 10,7 | 2,0 | | | | 0,9 | | 1,7 |
| 49 | | | | | | | 7,3 | 4,7 | 1,5 | | 0,6 | |
| 51 | | | | | 11,2 | 2,1 | | | | | | |
| 67,8 | 0,5 | 0,7 | 2,0 | 1,5 | | | | | | | 0,1 | 1,6 |
| 69 | | | | | | | 3,4 | 5,3 | 1,2 | | 0,1 | |
| 71,7 | 0,5 | 0,6 | 2,2 | 1,4 | | | | | | | | 0,9 |
| 73 | | | | | 11,6 | 1,4 | | | | 1,7 | | |
| 75 | | | | | 11,7 | 1,8 | | | | | | |
| 75,2 | | | | | | | 2,5 | 4,9 | 1,3 | | 0,2 | |
| 98,5 | | | | | | | 1,6 | 5,6 | 1,4 | 1,0 | | 0,7 |
| 112 | 0,5 | 0,7 | 3,0 | 1,5 | | | | | | | 0,2 | 0,6 |
| 125 | | | | | | | 0,7 | 2,5 | 1,5 | | | |
| 136 | 0,5 | 1,0 | 2,4 | 1,7 | | | | | | | | |
| 146 | | | | | | | 0,7 | 3,8 | 1,0 | 0,6 | | 0,3 |
| 148 | | | | | | | 0,4 | 3,3 | 1,2 | 0,6 | | 0,2 |
| 185 | 0,5 | 0,7 | 2,5 | 1,7 | | | | | | | | 0,3 |

Series F: Nitrogen and Simfuel pellet of burnup 30 MWd/kg U

Series L: Nitrogen and Simfuel pellet of burnup 30 MWd/kg U

Series N: Nitrogen and Simfuel pellet of burnup 50 MWd/kg U

TABLE A.5.- Flow experiments in GW water (ppb)

| t (h) | Mo ^G | Y ^G | Ba ^G | Sr ^G | Ba ^{CB} | Sr ^{CB} | Ba ^{CM} | Sr ^{CM} | U ^G | U ^{CB} | U ^{CM} |
|-------|-----------------|----------------|-----------------|-----------------|------------------|------------------|------------------|------------------|----------------|-----------------|-----------------|
| 2 | 0,6 | 0,9 | 2,5 | 2,1 | | | | | | 0,5 | |
| 4 | 0,5 | 0,8 | 2,3 | 1,8 | | | | | 1,9 | | |
| 6 | 0,5 | 0,7 | 2,4 | 1,9 | | | | | | | 1,2 |
| 8 | 0,5 | 0,7 | 2,2 | 1,9 | | | | | 1,8 | | |
| 11 | 0,5 | 0,7 | 2,3 | 1,8 | | | | | | 0,9 | |
| 13,5 | 0,5 | 0,6 | 2,3 | 1,8 | | | | | 1,3 | | |
| 24 | | | | | 1,6 | 2,5 | | | | | 1,1 |
| 25,5 | 0,5 | 0,6 | 2,3 | 1,8 | | | | | 0,8 | | |
| 28,5 | 0,5 | 0,6 | 2,3 | 1,7 | | | | | 1,8 | | |
| 29 | | | | | 1,8 | 2,7 | | | 1,1 | | |
| 29,5 | | | | | | | 1,7 | 2,4 | | 0,6 | |
| 31 | 0,5 | 0,7 | 2,3 | 1,7 | | | | | 0,9 | | |
| 33 | 0,5 | 0,8 | 2,3 | 1,8 | | | | | 1,3 | | |
| 35 | 0,5 | 0,8 | 2,3 | 1,8 | | | | | | 0,8 | |
| 50 | 0,5 | 0,6 | 2,4 | 1,4 | | | | | | | 1,4 |
| 52,5 | 0,5 | 0,5 | 2,3 | 1,7 | | | 2,0 | 2,3 | 2,1 | | |
| 54 | | | | | 2,1 | 2,7 | | | 0,5 | | |
| 55 | 0,5 | 0,4 | 2,3 | 1,3 | | | | | 1,9 | | |
| 57,5 | 0,5 | 0,4 | 2,2 | 1,4 | | | | | 0,4 | | |
| 76 | | | | | 2,6 | 2,7 | | | 0,4 | | 1,7 |
| 76,5 | | | | | | | 2,3 | 2,3 | | 0,6 | |
| 77 | 0,5 | 0,4 | 2,4 | 1,4 | | | | | 0,4 | | |
| 78 | | | | | 2,4 | 2,7 | | | 0,4 | | |
| 98,5 | | | | | 2,9 | 2,7 | | | | 0,6 | |
| 100 | | | | | | | 2,3 | 2,4 | | | 3,9 |
| 124 | 0,5 | 0,4 | 2,4 | 1,4 | 1,7 | 2,3 | | | 0,3 | | |
| 150 | | | | | 2,1 | 2,3 | | | | 0,5 | |
| 170 | | | | | | | 1,3 | 2,2 | | | 2,3 |
| 171 | | | | | | | 1,4 | 2,1 | | 1,2 | |

Series G: O₂/1%CO₂ with SIMFUEL pellet of burnup 30 MWd/kg U

Series CB: O₂/1%CO₂ with SIMFUEL pellet of burnup 50 MWd/kg U

Series CM: O₂/2%CO₂ with SIMFUEL pellet of burnup 50 MWd/kg U

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