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IN VITRO DISSOLUTION OF URANIUM PRODUCT SAMPLES FROM FOUR URANIUM MILLS

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ABSTRACT

The study was conducted to determine the rate of dissolution of uranium product (yellowcake) obtained from four uranium ore processing mills. Thirty day dissolution experiments were conducted *in vitro* using two solvents; a simulant of an ultrafiltrate of blood serum (SUF) containing diethylenetriaminepentaacetic acid (DTPA) and 0.1M HCl. Dissolution data are expressed as graphs of the logarithm of the percentage initial U undissolved vs time in hours and fitted to a negative exponential equation. The samples were characterized using x-ray powder diffraction and infrared spectroscopic techniques. All samples were mixtures of $(\text{NH}_4)_2 \text{U}_2\text{O}_7$ (ammonium diuranate) and $\alpha\text{-U}_3\text{O}_8$. The combined results show that $(\text{NH}_4)_2 \text{U}_2\text{O}_7$ dissolved much more rapidly in either solvent than does $\alpha\text{-U}_3\text{O}_8$. Dissolution half times in SUF containing DTPA were: $(\text{NH}_4)_2 \text{U}_2\text{O}_7 \approx 10$ hr, $\alpha\text{-U}_3\text{O}_8 \approx 10^4$ hr. The percentage of total material present as the more soluble $(\text{NH}_4)_2 \text{U}_2\text{O}_7$ form was shown to vary from 99.4% to 51%. The results indicate that caution must be exercised in the interpretation of bioassay results of workers from different U mills since the highly variable chemical composition of yellowcake may result in widely variable *in vivo* solubility and excretion.

Introduction

A study was conducted to determine the rate of solubility of uranium product (yellowcake) obtained from four uranium processing mills. The techniques employed in the study were developed in this Institute to assess the solubilization rate of finely divided particulates in various solvents. The primary solvent employed in this study was a simulant of an ultrafiltrate of blood serum,¹ SUF, containing DTPA. Such studies in an *in vitro* system should be valuable in predicting the potential solubilization that would occur *in vivo* following inhalation of the particulate material in humans. A second solvent, 0.1 M HCl, was employed on duplicate samples to assist in elucidating chemical mechanisms operative in the solubilization process. The duration of the study was 30 days. An adjunct to the *in vitro* solubilization study was the determination of chemical form of the uranium in each sample using x-ray powder diffraction measurements. Infrared spectroscopy was also employed on some samples of yellowcake to better characterize some relatively non-crystalline forms of uranium that were present.

Materials and Methods

Approximately 3 grams of yellowcake powder were obtained from the production lines of each of four uranium mills. Details of previous process history or sample collection were unknown. The samples were identified only by the name of the company operating the mill.

A 25-29 mg portion of each sample was weighed on a Nucleopore filter membrane (pore size 0.1 μm) and a second filter of identical type was placed over the sample. The filters containing the powder sample were secured by compression of an O-ring near their periphery by a circular teflon clamp. This assembly was then placed in 200 ml of solvent.² This *in vitro* system allowed for complete retention of the particulates between the two filters while allowing free diffusion of the solvent and solute. The solvent was changed every two hours for the first six hours. Beyond this time through 30 days, solvent was changed periodically as shown in Figures 1 and 2.

IN VITRO SOLUBILITY OF YELLOWCAKE IN SERUM SIMULANT + DTPA

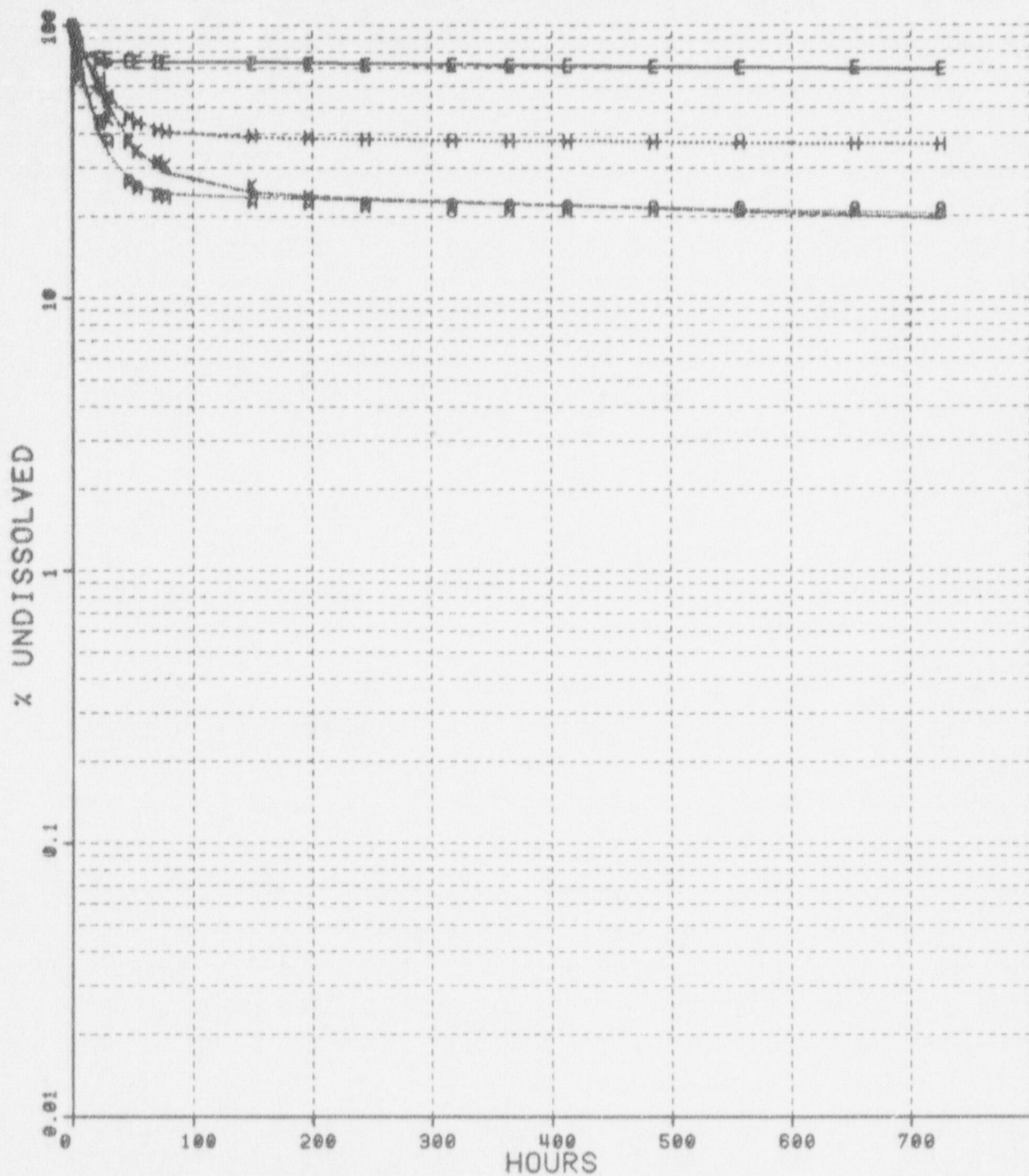


Figure 1. *In vitro* dissolution of yellowcake obtained from four uranium mills in serum simulant containing DTPA. E, Exxon Mill; H, Homestake Mill; K, Kerr-McGee Mill; A, Anaconda Mill.

IN VITRO SOLUBILITY OF YELLOWCAKE
IN 0.1M HCL SOLVENT

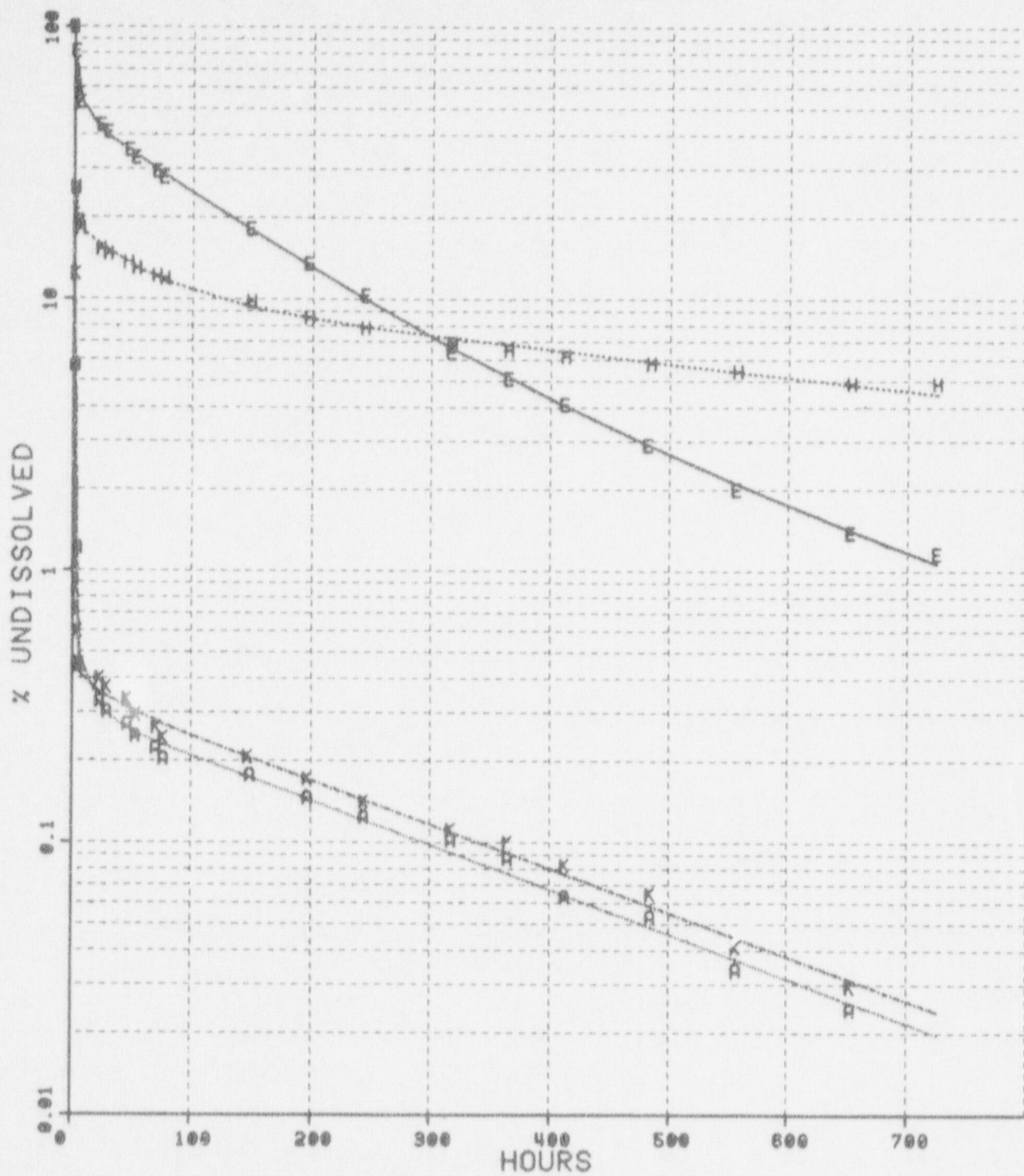


Figure 2. *In vitro* dissolution of yellowcake obtained from four uranium mills in 0.1 M HCl. E, Exxon Mill; H, Homestake Mill; K, KerrMcGee Mill; A, Anaconda Mill.

The composition of the SUF used in these studies is detailed in Table 1. The second solvent employed with duplicate samples was 0.1 M HCl.

Table 1
Composition of Simulated Serum Ultrafiltrate (SUF) Used
to Study the Dissolution of Yellowcake Samples
(pH = 7.6)

| <u>Salt</u> | <u>Molar Concentration</u> |
|---|--------------------------------|
| NaCl | 0.116 |
| NH ₄ Cl | 0.010 |
| NaHCO ₃ | 0.027 |
| Glycine | 0.005 |
| Na ₃ Citrate·2H ₂ O | 0.0002 |
| CaCl ₂ ·2H ₂ O | 0.0002 |
| L-cystine | 0.001 |
| H ₂ SO ₄ | 0.0005 |
| Na ₂ HPO ₄ ·7H ₂ O | 0.0012 |
| DTPA ^a | 0.0002 |

^aNot present in blood serum. Alkylbenzyltrimethylammonium chloride (2 ml) added as an antibacterial agent.

Analysis for uranium content of each solvent sample and the material remaining in the filter sandwich at the end of the 30-day experimental period was accomplished by fluorimetry after fusion of an aliquot in a NaF-LiF salt mixture.³ Appropriate standard solutions and blank samples were employed to ensure accuracy and precision of the uranium determinations. The initial quantity of uranium present was determined by summing the total uranium in each solvent sample plus the quantity measured in the filter sandwich at conclusion of the study. This value plus the periodic measurements of dissolved uranium in the solvent were used to express the results as a percentage undissolved as a function of time in the solvent. The data, plotted as the percent of uranium undissolved versus time, were fitted using a non-linear least squares computer program with a two or three-component negative exponential equation of the form

$$\% \text{ undissolved} = A_1 e^{-\lambda_1 t} + A_2 e^{-\lambda_2 t} + \dots + A_n e^{-\lambda_n t}$$

where t in hours is the elapsed time of the study. The fractions of the total sample dissolved, A_i , and the corresponding dissolution half-time, λ_i , associated with each component of these fitted curves, were obtained.

Materials were analyzed using a Phillips APD-3501 automated x-ray powder diffractometer to determine the chemical form of the uranium present. Specimens of bulk samples were prepared according to standard procedures with no grinding or other pretreatment. Additional specimens of materials which had been subjected to dissolution for two hours were removed from the filter sandwich holder and dried at 50°C for two days. The dried specimens were scraped from the membrane filter and were sifted through a wire screen onto an uncoated, silver membrane filter of 0.1 μm pore size. This technique deposited an even coating of powder for x-ray diffraction analysis.

Infrared spectroscopic analysis was used to study specimens which were of low crystallinity or were present in such small relative amounts that x-ray diffraction studies were not adequate. Specimens for infrared spectroscopy were prepared using standard KBr disk or nujol mull techniques.

Results

The four samples of yellowcake supplied were finely divided powders of unknown particle size distribution. The four powders were quite different in appearance as summarized in Table 2.

Table 2
Visual Observations Made During the First
Twenty-Four Hours of Dissolution Study

| <u>Source</u> | <u>Initial Color</u> | <u>Final Color</u> | |
|---------------|----------------------|--------------------|----------------------------------|
| | | <u>0.1 M HCl</u> | <u>SUF^a</u> |
| Anaconda | dull yellow | brown | dull yellow |
| Kerr-McGee | bright yellow | no sample visible | bright yellow ≈ 1/2 dissolved |
| Homestake | brown/green | gray/black | brown/green |
| Exxon | black/green | black | black with yellow diffused |

^aSee Table 1.

At early times in the dissolution study, changes were noted in the color of the particulate material retained in the filter sandwich. These color changes were different for the two solvents. These observations are summarized in Table 2. After the first 24 hours in solution, the appearance of the particulate material did not change through the 30-day experimental period.

The results of the dissolution studies in the SUF are presented graphically in Figure 1. The fraction of material dissolved and corresponding halftimes of dissolution expressed in hours are tabulated in Table 3. Results for the studies in 0.1 M HCl are summarized in Figure 2 and Table 4, respectively.

Table 3

Calculated Values for Percent of Total Material Remaining and Half-times of Dissolution for Four Uranium Product (Yellowcake) Samples as Determined in *in Vitro* Dissolution Studies Using Serum Simulant + DTPA as Solvent

| <u>Uranium Mill</u> | <u>%^a</u> | <u>T_{1/2}</u> <u>(hours)</u> | <u>%</u> | <u>T_{1/2}</u> <u>(hours)</u> | <u>%</u> | <u>T_{1/2}</u> <u>(hours)</u> |
|---------------------|----------------------|--|----------|--|----------|--|
| Anaconda | 24.9 | 2.4 | 53.1 | 16.1 | 22.0 | 16,900 |
| Kerr-McGee | 63.6 | 14.2 | 16.4 | 85.1 | 20.0 | 13,000 |
| Homestake | 60.9 | 16.0 | | | 39.1 | 7,800 |
| Exxon | 25.5 | 3.4 | | | 74.5 | 6,600 |

^aAll values obtained by curve fit of data using a negative exponential equation.

Table 4

Calculated Values for Percent of Total Material Remaining and Half-times of Dissolution for Four Uranium Product (Yellowcake) Samples as Determined in *in Vitro* Dissolution studies Using 0.1 M HCl as Solvent

| <u>Uranium Mill</u> | <u>%</u> | <u>T_{1/2}</u> <u>(hours)</u> | <u>%</u> | <u>T_{1/2}</u> <u>(hours)</u> | <u>%</u> | <u>T_{1/2}</u> <u>(hours)</u> |
|---------------------|----------|--|----------|--|----------|--|
| Anaconda | 99.4 | 0.5 | 0.3 | 8.6 | 0.3 | 183 |
| Kerr-McGee | 99.5 | 0.5 | 0.06 | 23.0 | 0.4 | 186 |
| Homestake | 81.1 | 0.6 | 8.6 | 40.0 | 10.3 | 600 |
| Exxon | 51.0 | 2.1 | 38.3 | 85.5 | 10.7 | 208 |

Results of x-ray powder diffraction studies of the four yellowcake samples are shown in Figures 3-6 and are summarized in Table 5. The assignments of the x-ray diffraction peaks to alpha- U_3O_8 and $(NH_4)_2U_2O_7$ (ammonium diuranate) were made by comparison with the diffraction pattern of known alpha- U_3O_8 and with the diffraction pattern of $(NH_4)_2U_2O_7$ prepared in the laboratory. Where $(NH_4)_2U_2O_7$ was of low crystallinity or present in trace amounts, infrared spectroscopy was used to confirm its presence.

Table 5

Qualitative Analysis of Yellowcake Samples From Four Uranium Mills

| <u>Industrial Mill</u> | <u>Alpha-U_3O_8 Present</u> | <u>$(NH_4)_2U_2O_7$ Present</u> |
|------------------------|--|--|
| Anaconda | - | + ^a |
| Kerr-McGee | - | + |
| Homestake | + | + |
| Exxon | + | + ^a (trace) |

^aPoorly crystalline or minor fraction; presence confirmed by infrared spectroscopy.

X-ray diffraction patterns were also obtained from powders which had been subjected to one of the two solvents used. Representative diffraction patterns for the Homestake sample after contact with each solvent are shown in Figure 7. Comparison between these two patterns and the materials corresponding with the x-ray diffraction pattern prior to dissolution (Figure 5), shows that in SUF containing DTPA, the $(NH_4)_2U_2O_7$ dissolved somewhat more rapidly than U_3O_8 in two hours, while in 0.1 M HCl the $(NH_4)_2U_2O_7$ dissolved completely leaving only U_3O_8 . The general results seen with the other materials were that the Anaconda and Kerr-McGee samples dissolved in both solvents while retaining their identity as $(NH_4)_2U_2O_7$. The Exxon sample also retained its identity upon dissolution.

Discussion

The variation in the appearance of the four yellowcake samples indicated that more than one chemical form of uranium was present among the four samples. Uranium sesquioxide (U_3O_8) is olive green to black in color while ammonium

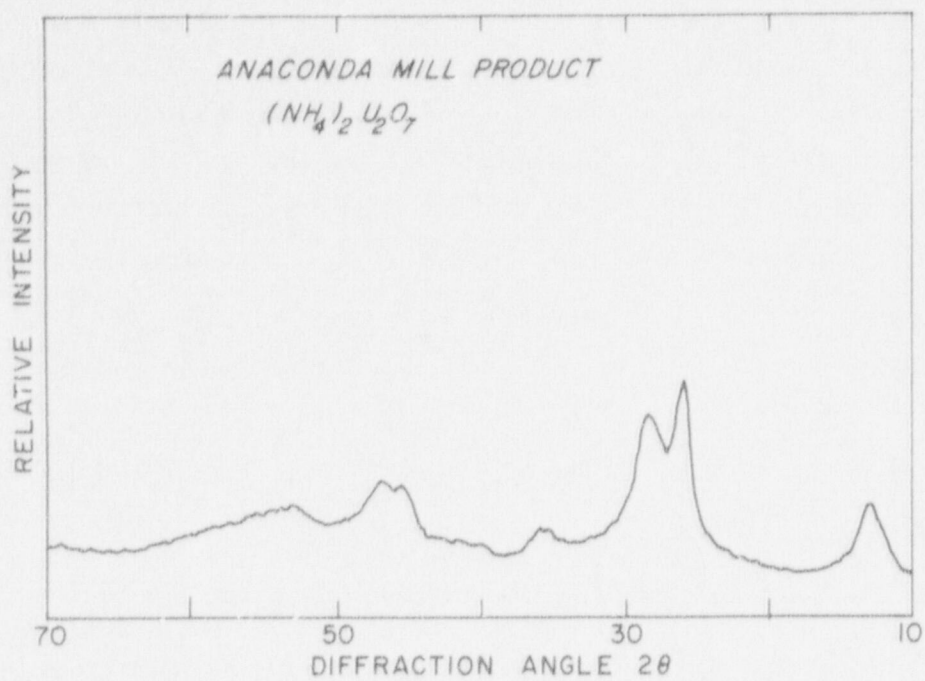


Figure 3. X-ray diffraction pattern of bulk yellowcake from Anaconda Mill; primarily ammonium diuranate.

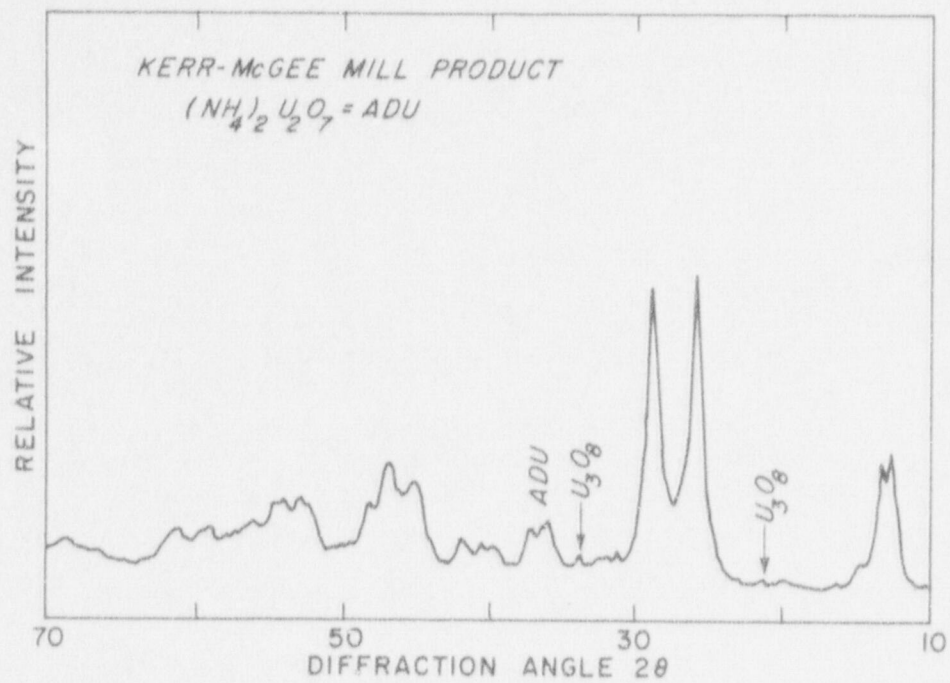


Figure 4. X-ray diffraction pattern of bulk yellowcake from Kerr-McGee Mill; primarily ammonium diuranate.

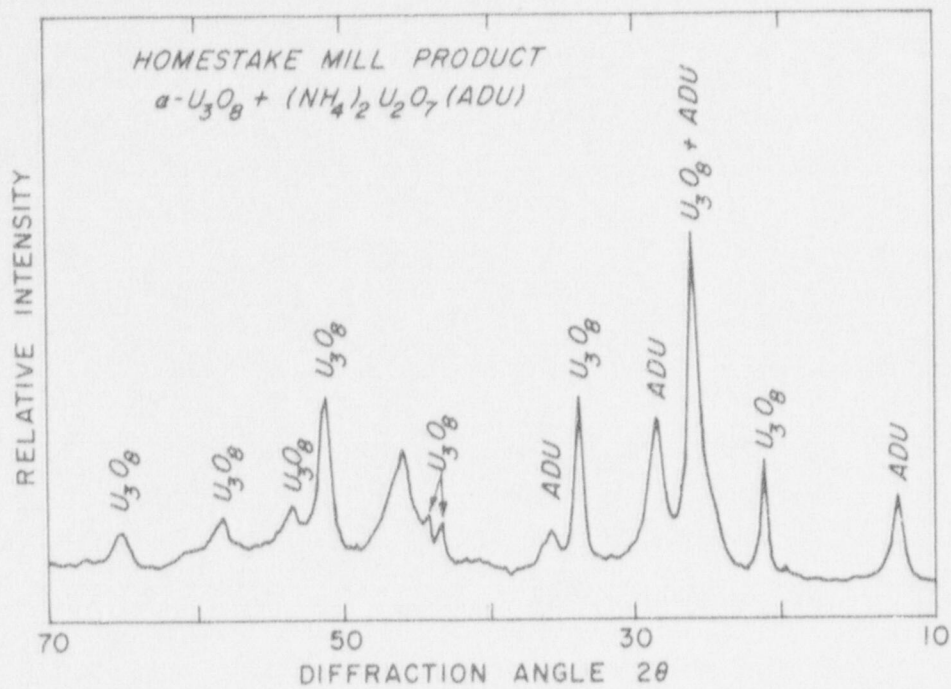


Figure 5. X-ray diffraction pattern of bulk yellowcake from Homestake Mill; mixture of U_3O_8 and ammonium diuranate.

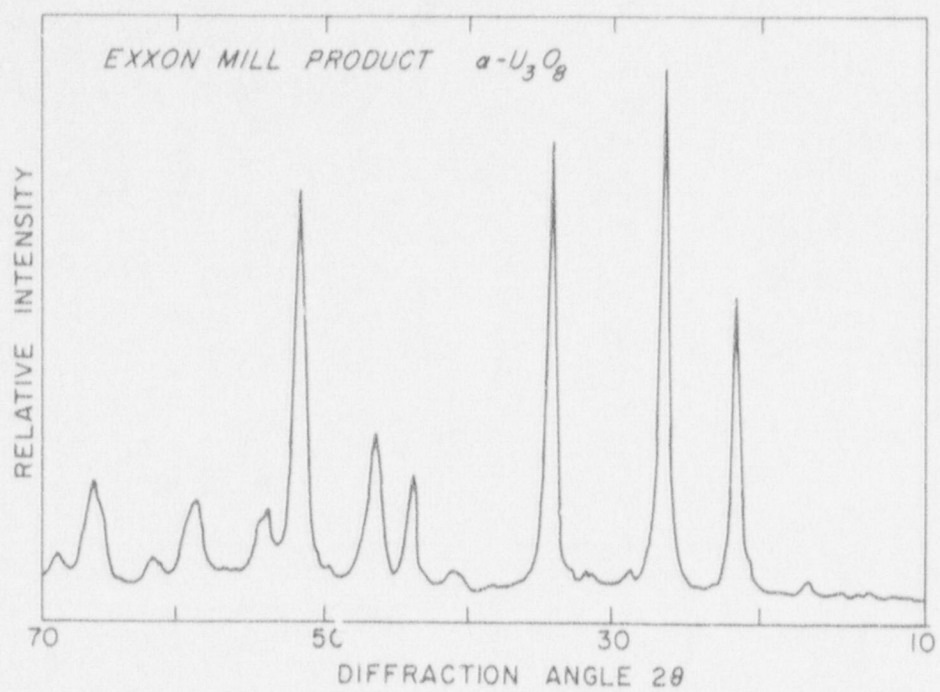


Figure 6. X-ray diffraction pattern of bulk yellowcake from Exxon Mill; primarily U_3O_8 .

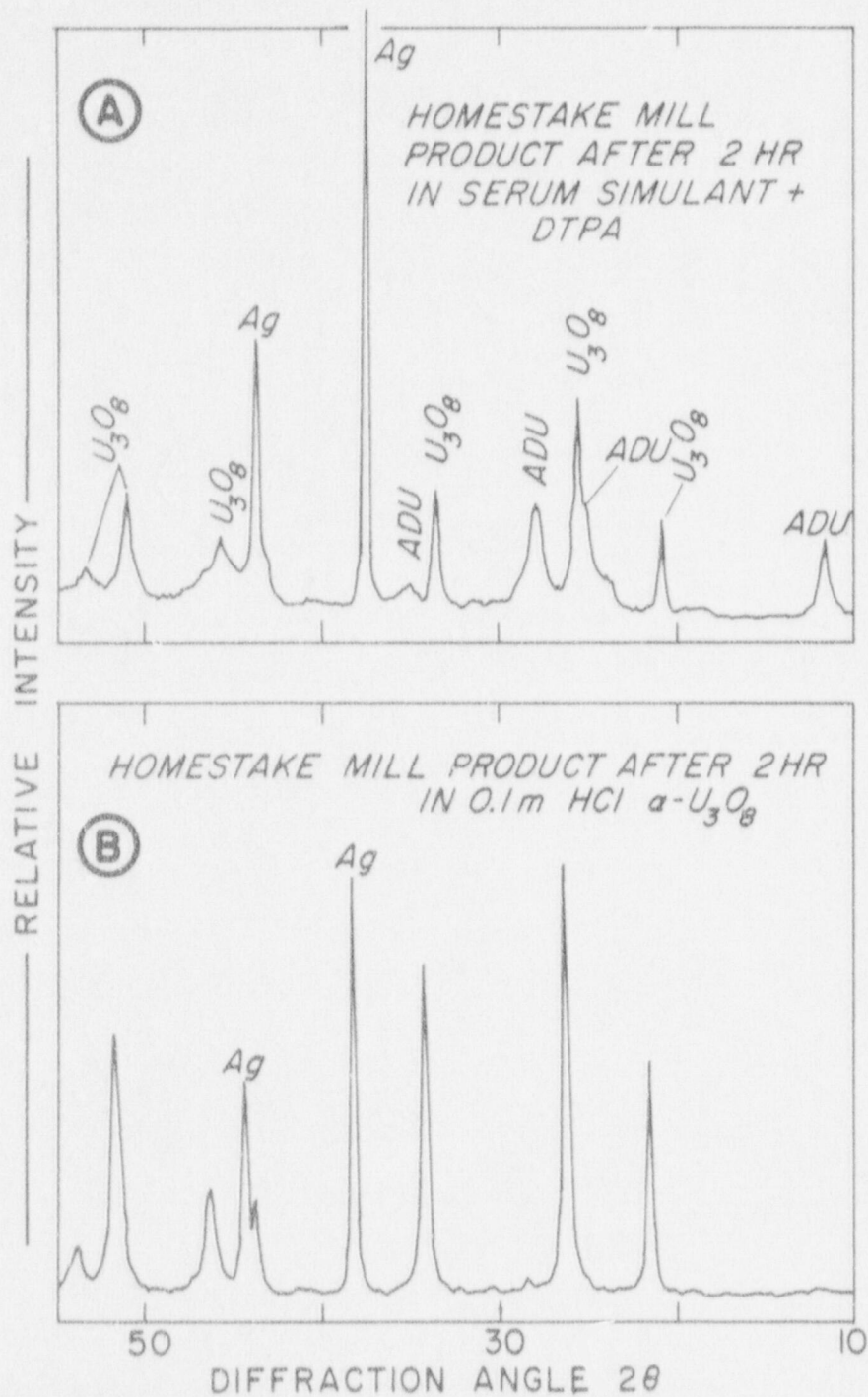


Figure 7. X-ray diffraction pattern of bulk yellowcake from Homestake Mill after two hour dissolution in (A) serum simulant with DTPA and (B) 0.1 M HCl. Ag = diffraction peak due to silver membrane substrate used as calibration standard.

diuranate $[(\text{NH}_4)_2\text{U}_2\text{O}_7]$ is yellow. Thus, at the onset of this work it was suspected that the samples from Exxon and Homestake were predominantly U_3O_8 while the Kerr-McGee and Anaconda samples were predominantly ammonium diuranate. The color changes in the sample trapped in the filter holder noted during the first six hours of dissolution indicated that two chemical forms were potentially present in all samples.

Confirmation of the presence of at least two chemical forms of uranium was obtained from the powder x-ray diffraction measurements. The Exxon sample was determined to be primarily alpha- U_3O_8 with a small amount of ammonium diuranate present. The Homestake sample was a mixture of alpha- U_3O_8 and ammonium diuranate. The samples from Kerr-McGee and Anaconda were quite similar, being primarily ammonium diuranate with a small amount of another form of uranium present. However, these latter two samples differed in the degree of crystallinity of the ammonium diuranate. The Anaconda sample was less crystalline than the Kerr-McGee sample as may be seen by comparing Figures 3 and 4. In the Anaconda and Exxon samples where the ammonium diuranate was of low crystallinity or present in a small amount, its presence was confirmed by infrared spectroscopy.

In general, the results of *in vitro* dissolution studies show that the dissolution rates for all materials were greater in 0.1 M HCl than in the SUF containing DTPA. The results shown in Figure 7 for the x-ray diffraction patterns of materials which had been subjected to dissolution for two hours show that $(\text{NH}_4)_2\text{U}_2\text{O}_7$ was the more soluble component. The physical significance of the two-component dissolution rate curves is that the first, more rapid component was due primarily to the dissolution of the $(\text{NH}_4)_2\text{U}_2\text{O}_7$ fraction, while the slower component was due primarily to the remaining U_3O_8 fraction. However, several of the dissolution rate curves are best described by a three-component equation. Inspection of the data in Tables 3 and 4 shows that the $T_{1/2}$ values of the third components of the rate curves for each material in a given solvent are similar, and the same comparison is true for the $T_{1/2}$ values first components, leading to the conclusion that the first component represents the dissolution rate of $(\text{NH}_4)_2\text{U}_2\text{O}_7$ while the third component is due to U_3O_8 dissolution. The second component is more difficult to assign, since there appear to be only two predominant chemical forms of uranium oxide present. However, it is possible

that the second component may represent the result of the very wide range of particle sizes present in the samples received. The second component could include dissolution of large $(\text{NH}_4)_2\text{U}_2\text{O}_7$ particles or very small U_3O_8 particles.

A more quantitative determination of the relative amounts of alpha- U_3O_8 and $(\text{NH}_4)_2\text{U}_2\text{O}_7$ present can be made using the results of the *in vitro* dissolution studies shown in Tables 3 and 4. The results of the studies using SUF containing DTPA as a solvent (Table 3) show that the Exxon and Homestake samples can be described by two-component exponential functions whereas the samples from Kerr-McGee and Anaconda require three-component exponential equations. The dissolution of powder from Exxon is characterized by 25% of the uranium dissolving with a half-time of 3.4 hours followed by 75% of the material dissolving with a half-time of 6600 hours (\approx 275 days). The Homestake powder, while showing the same biphasic dissolution, is characterized by 61% of the material dissolving with a half-time of 16 hours followed by 39% of the material dissolving with a halftime of 7820 hours (325 days). The dissolution of powders from Kerr-McGee and Anaconda required three-component exponential equations to adequately describe the data. The initial phase of dissolution of the Kerr-McGee sample involved 64% of the material dissolving with a half-time of 14.2 hours followed by an intermediate phase with 16% of the material dissolving with a half-time of 85 hours and a long term component in which 20% of the material dissolved with a half-time of 13,000 hours (540 days). Similarly, the Anaconda sample dissolved with a first component of 25% of the material (half-time \approx 2.4 hours), a second component of 53% of the material (half-time \approx 16 hours), followed by a third component of 22% of the material (16,900 hours \approx 700 days). In the case of the Kerr-McGee and Anaconda samples, it was interesting to note that the long-term component was represented by nearly identical quantities of material and with half-times that were not statistically different, such that, by 150 hours after dissolution began, the dissolved fractions and half-times of the two materials appeared identical. The dissolution half-time for this third component (275-700 days) agrees with the dissolution constant determined in cases of human or animal exposures to U_3O_8 (140-500 days).⁴ This supports the assignment of the long-term component to the dissolution of U_3O_8 in these yellowcake samples.

From these data obtained in SUF, it is apparent that 25 to 64% of each sample of yellowcake tested showed a rapid initial dissolution with half-times of less than 16 hours. When the intermediate components for the Kerr-McGee and Anaconda samples were added to the respective initial components, the results indicate that $\approx 80\%$ of both materials dissolved within ≈ 200 hours. The dissolution curves of the Kerr-McGee and Anaconda samples show that this 20% of the material remaining dissolved with a half-time which was too long to be associated with $(\text{NH}_4)_2\text{U}_2\text{O}_7$ although x-ray diffraction and infrared spectroscopic results indicated only $(\text{NH}_4)_2\text{U}_2\text{O}_7$ to be present. One possible explanation for this is that since the SUF was buffered at pH 7.6, the dissolved uranium was present as a hydroxide which was less soluble than $(\text{NH}_4)_2\text{U}_2\text{O}_7$ and was dissolved at a slower rate. The exact magnitude of the long-term components of dissolution should be viewed with some skepticism due to the very long predicted half-times compared to the length of the study (30 days).

The dissolution of duplicate samples from each uranium mill in the 0.1 M HCl solvent required three-component exponential equations to adequately describe the data. The Kerr-McGee and Anaconda samples were not statistically different from each other and showed nearly complete dissolution within the 30-day study period. Initially, the fraction dissolved and the half-time of dissolution were in the same order among the four samples as measured in the SUF. In the third dissolution phase, the Exxon sample showed that a comparable fraction dissolved with a shorter half-time than did the Homestake sample in contrast to the data obtained in the serum simulant.

This difference may be due to other powder characteristics not considered here. Additional factors of potential importance in determining the solubility of the uranium product include (1) the particle size distribution of the powder, (2) more complete identification of the chemical species present initially. The role of factors such as (1) the variation, if any, in the chemical forms of U present from batch to batch from any given uranium mill and (2) identification of any potential intermediate chemical form formed during dissolution as determinants of the *in vivo* behavior of deposited uranium should be carefully assessed in order to validate protection standards and bioassay procedures.

Within the limitations noted above, the results of these dissolution studies indicate that:

1. Significant variation exists in the *in vitro* solubility of the product (yellowcake) from four uranium mills.
2. These variations in solubility may be related to differences in the chemical species present in the product.
3. These results indicate that caution must be exercised in the interpretation of bioassay results of workers from different uranium mills because the different chemical forms present may yield different results.
4. Additional factors, not within the scope of this study, may also be important in the solubility of these materials.

Results summarized in this report have shown that in bulk yellowcake samples supplied by different uranium mills, the composition varied from > 99% highly soluble $(\text{NH}_4)_2\text{U}_2\text{O}_7$ (Anaconda, Kerr-McGee) to approximately 25% $(\text{NH}_4)_2\text{U}_2\text{O}_7$ + 75% less soluble U_3O_8 . Determination of the particle size distribution would aid in determining the amount of respirable dust present and the concentration of either $(\text{NH}_4)_2\text{U}_2\text{O}_7$ or U_3O_8 in the respirable fraction.

Several forms of ammonium diuranate are known which are related by the relative amounts of nitrogen and uranium present. The determination of the N/U ratio for yellowcake samples would allow a more complete identification of the yellowcake samples using x-ray diffraction or infrared techniques, and would be a measure of the reproducibility of batch processes for a given mill. A reliable rapid technique to determine the N/U ratio would serve as a guide to the industrial health officer at a uranium mill in his determination of the relative solubility or insolubility of the mill product for a given batch.

Identification of the chemical species in solution during *in vitro* dissolution studies would aid in their interpretations as well as aid in the development of a solvent which would relate more closely to the biological behavior of mill products. This is especially true in the case of ammonium diuranate which has not been as extensively studied in biological systems as has U_3O_8 .

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