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DISSOLUTION FRACTIONS AND HALF-TIMES OF SINGLE SOURCE YELLOWCAKE IN SIMULATED LUNG FLUIDS*

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Abstract—Thirty-five samples of yellowcake, the primary product of uranium milling, were obtained from industry. Samples were selected from the archives of 6 yr of yellowcake production from a single uranium milling operation. Ten representative samples were selected on the basis of color variation and chemical composition. Dissolution fractions and half-times were obtained using both simulated interstitial and simulated surfactant lung fluids maintained in vials submerged in a shaker water bath at 37°C for 26 days. Samples were collected at 12 different time intervals and fresh simulant replaced. Uranium concentrations were analyzed by fluorometric methods using ASTM techniques. The fraction of the uranium undissolved, F, can be well modelled by the sum of three exponential terms:

$$F = f_1 \cdot \exp\left[\frac{-0.693t}{t_{1/2}(1)}\right] + f_2 \cdot \exp\left[\frac{-0.693t}{t_{1/2}(2)}\right] + f_3 \cdot \exp\left[\frac{-0.693t}{t_{1/2}(3)}\right],$$

with $f_1 + f_2 + f_3 = 1$.

Dissolution fractions and half-times in the two simulated lung fluids were found to be not significantly different. The point estimates for the six parameters (three fractions of the amount of uranium and the three associated half-times for dissolution) based on the analyses are: f_1 equals 16.0% and $t_{1/2}(1)$ equals 3.5 hr, f_2 equals 16.5% and $t_{1/2}(2)$ equals 4.4 days, and f_3 equals 67.5% and $t_{1/2}(3)$ equals 224 days. The final characterization of yellowcake resulting from this investigation strongly suggests that dissolution rates of yellowcake from a single source may be individually process specific or site specific.

INTRODUCTION

YELLOWCAKE is a generic term referring to the primary end product of uranium milling

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which can vary in composition from ammonium diuranate to mixtures of uranium oxides, sulfates, nitrates and sodium compounds. Experimental work showed uranium in yellowcake to be more soluble in lung fluids than previously reported (Ka78; Ei78). These two investigations utilized splits of four yellowcake samples supplied by one of the conversion facilities. The samples obtained were assumed to be representative of the

yellowcake milling industry. It was not known by either investigator that different chemical extraction techniques or drying temperatures existed among the randomly selected yellowcake products. Although some variability in dissolution fractions and half-times among the four samples investigated was shown by both investigators, only one investigator felt the differences to be significant enough to warrant caution in describing a blanket solubility characterization for the milling products investigated (Ei78). Dissolution half-time is defined as the time required for one half of the deposited material to solubilize in the lung. These results led the Nuclear Regulatory Commission (NRC) to change its position regarding the solubility classification of uranium in yellowcake for the lung from an insoluble (Y) classification (greater than 100-day dissolution half-time) to a mixed classification including a soluble (D) classification (less than 10-day dissolution half-time) component, a less soluble (W) classification (10- to 100-day dissolution half-time) component, and a Y component (A178). Data available to NRC indicated that each yellowcake sample investigated contained a significant soluble fraction (Ka78; Ei78). The NRC has further asserted this viewpoint in the pbulication of Regulatory Guide 8.22 (For Comment) which outlines an acceptable bioassay program for uranium mills (Nu78).

A blanket classification of yellowcake implies that all uranium compounds referred to as yellowcake exhibit very similar behaviors. However, visual examination of 35 industrial yellowcake samples demonstrated large color variations and suggested that yellowcake composition varies widely among different production runs (De79). All samples collected for this investigation were obtained from the archives of a single acid-leach, uranium milling operation which utilizes a high-temperature calcination process. The 10 representative yellowcake samples used in this investigation varied from bright yellow to dark mossy green and uranium content varied from 78.7 to 81.84%. Other chemical characteristics also varied as can be seen in Table 1. The density was fairly uniform, ranging from

5.7 to 7.2 g/cm³. The particle size ranged from 1.4 to 4.8 μm, and the surface area ranged from 9.2 to 23.8 m²/g (see Table 1). This type of variation was also demonstrated by Kalkwarf; color varied from yellow to black, uranium content varied from 71.67 to 78.60%, ammonia varied from 0.32 to 2.6%, and sulfate varied from 1.07 to 6.57% (Ka78).

Previous research by Moss (Mo77) Kalkwarf (Ka78), Cooke and Holt (Co74) and Steckel and West (St66) examined the solubility of various uranium compounds in simulated interstitial lung fluid. Since surfactant lung fluid is believed to be present at the air–liquid interface in the alveoli, its effect upon dissolution of an inhaled particle should be defined. The primary surface active component found in all mammalian lungs studied is dipalmitoyl lecithin, DPL (Mu76). This investigation utilizes both simulated surfactant and simulated interstitial lung fluids.

METHODS AND MATERIALS

Difficulties in obtaining actual lung fluids in large quantities have been recognized by King and Clements (Ki72). Therefore, the simulated lung fluids must model original solutions in the lungs closely in ionic composition, pH, and general conditions (Mo79). Table 2 shows that the prepared simulants closely resembled the constituents of human lung fluid as analyzed by Diem and Lentner (Di70). The simulated lung fluids were prepared according to modifications of Gamble's solution (Ga54; Ka78; Mo79). Citrate was substitued for protein due to probable bacterial action. Organic acid anions were replaced with acetate. Simulated surfactant lung fluid differs from simulated interstitial lung fluid by the addition of 200 μg/ml DPL (A179).

Two weighed quantities of each yellowcake sample, ranging from 300 to 450 mg, were placed into separate polyethylene vials. Twenty ml of one of the simulated fluids was added to each vial. Vials were capped, hand-shaken, and submerged in an ultrasonic water bath for 5 min to disperse any agglomerates. Vials were transferred to a shaker water bath, maintained at 37°C, and agitated continuously to approximate human lung conditions and

Table 1. *Physical and chemical characteristics for yellowcake samples*

	1	2	3	4	5	6	7	8	9	10	Average ± 2σ
Physical Characteristics											
Density (g/cm³)	6.21	5.68	6.70	6.87	6.87	6.92	7.12	7.04	7.23	7.23	6.79 ± .98
Particle size (ESD_{50}, μm)	2.9	2.7	2.1	2.2	1.4	2.2	2.1	3.2	3.8	4.8	2.7 ± 1.9
Surface area (m²/g)	20.1	18.2	23.8	15.6	10.9	9.2	15.6	15.5	13.4	13.2	15.6 ± 8.6
Chemical Characteristics*											
% U	78.7	79.3	81.3	80.69	80.7	81.71	81.84	81.33	81.52	81.42	
% NH_4	.63	.81	.14	.26	.17	.28	.12	.12	.04	.01	
% NO_3	<.02	<.02	<.02	.10	<.02	<.02	.23	<.02	<.02	<.02	
% SO_4	<3.0	<3.0	2.53	3.2	3.4	.89	2.88	<3.0	1.57	2.3	
% CO_3	<.2	<.2	.03	.03	.6	<.02	.03	.2	.02	.01	
% Cl	<.05	<.05	<.01	<.01	.02	<.05	<.01	<.05	<.01	<.01	
% F	<.01	<.01	<.01	<.01	<.002	<.01	<.01	<.01	<.01	<.01	
% Mo	<.01	<.01	<.01	<.01	<.01	<.01	<.01	<.01	<.01	<.01	
% V	<.1	<.1	.01	.01	.02	<.01	.02	<.1	.03	.02	
% Na	NA	NA	.08	.03	.06	NA	.07	NA	.08	.08	
% K	NA	NA	.02	.02	.08	NA	.01	NA	.03	.03	
% Ca	<.05	<.05	.05	.04	.04	.11	.04	<.05	.12	.12	
% Fe	NA	<.15	.09	.06	.23	.12	.07	NA	.42	.3	

NA: Not analyzed

*Chemical analyses were performed by several different laboratories with varying limits of detection for specific tests.

Table 2. *Composition of lung fluids*

Ions	Actual (Di70) (meq)	Simulated Interstitial (meq)	Simulated Surfactant (meq)
Calcium	5.0	5.0	5.0
Magnesium	2.0	2.0	2.0
Potassium	4.0	4.0	4.0
Sodium	145.0	145.0	145.0
Total Cations	156.0	156.0	156.0
Bicarbonate	31.0	31.0	31.0
Chloride	114.0	114.0	114.0
Citrate	--	1.0	1.0
Acetate	7.0	7.0	7.0
Phosphate	2.0	2.0	2.0
Sulfate	1.0	1.0	1.0
Protein	1.0	--	--
Dipalmitoyl Lecithin	--	--	200 μg/ml
Total Anions	156.0	156.0	156.0
pH	7.3	7.3	7.3

maximize dissolution rates. The pH of the lung fluids was maintained near 7.3 by addition of dilute HCl acid as the pH slowly drifted upward.

Samples were removed from the water bath at selected time intervals and centrijuged to separate any suspended particles from solution. The supernate was pipetted off the yellowcake, fresh simulant fluid replaced, and the entire treatment repeated. Samples were taken at 2, 4, 6, 8, 10, 36, 60, 84, 108, 180, 348 and 636 hr.

The supernate was analyzed for uranium by a slight modification of standard fluorometric techniques (Am75). Briefly, 500 μl of supernate was placed into a 40-ml glass vial, 2 drops 0.2 M KMnO₄ were added, followed by 1 drop 2.0 M NH₂OH·HCl. Ten ml of acid-deficient aluminum nitrate salting solution with tetrapropylammonium hydroxide was added to the sample followed by 5 ml of methyl iso-butyl ketone. The vial was shaken and 100 μl of sample was pipetted onto each of two platinum dishes containing 0.5 g 98% NaF-2% LiF powdered flux mixture. After a 15-min drying period under IR lamps, the dishes were placed onto a fusion ring burner, heated to 500°C for 30 sec, and fused at 950°C for 3.0 min. Uranium

concentrations were determined using a Jarrell–Ash fluorometer. Duplicate blanks and uranium standards were assayed with every fusion sample set (De79).

RESULTS AND DISCUSSION

The fluorometric analyses permit calculation of the fraction of uranium remaining undissolved in the yellowcake samples. Table 3 summarizes these results. In every case, the correlation coefficient of data to the generated regression fit is greater than 0.999. Non-linear regression analysis reveals that the dissolution is well described by the sum of three exponential terms with coefficients f_1, f_2 and f_3 representing that fraction of sample dissolving with dissolution half-times $t_{1/2}(1)$, $t_{1/2}(2)$ and $t_{1/2}(3)$, respectively (see equation 1).

$$F = f_1 \cdot \exp\left[\frac{-0.693t}{t_{1/2}(1)}\right] + f_2 \cdot \exp\left[\frac{-0.693t}{t_{1/2}(2)}\right]$$
$$+ f_3 \cdot \exp\left[\frac{-0.693t}{t_{1/2}(3)}\right], \qquad (1)$$

with $f_1 + f_2 + f_3 = 1$. F equals the fraction of uranium remaining undissolved in the sample at time t. A model that could have been fit to

Table 3. Fraction of uranium remaining undissolved at sampling intervals

						Time in Hours						
	2	4	6	8	10	36	60	84	108	180	348	636
1 I	.929	.896	.857	.823	.787	.746	.708	.667	.626	.561	.476	.383
1 S	.933	.886	.838	.785	.742	.698	.654	.609	.582	.535	.454	.360
2 I	.952	.923	.889	.858	.825	.794	.758	.726	.678	.621	.505	.398
2 S	.946	.911	.880	.837	.806	.767	.734	.705	.675	.624	.552	.460
3 I	.925	.896	.860	.823	.796	.777	.757	.735	.703	.652	.566	.470
3 S	.939	.921	.886	.857	.828	.804	.778	.751	.719	.674	.614	.541
4 I	.930	.903	.864	.829	.797	.770	.748	.726	.692	.633	.579	.518
4 S	.941	.912	.880	.845	.820	.796	.772	.740	.712	.666	.609	.552
5 I	.938	.918	.894	.869	.852	.833	.814	.796	.771	.734	.702	.665
5 S	.937	.916	.893	.868	.850	.831	.816	.800	.781	.742	.709	.674
6 I	.975	.954	.932	.910	.888	.869	.849	.829	.809	.776	.738	.700
6 S	.972	.949	.924	.897	.876	.853	.832	.811	.786	.759	.720	.683
7 I	.949	.915	.890	.867	.844	.823	.803	.780	.756	.724	.680	.624
7 S	.951	.914	.883	.849	.824	.801	.779	.756	.733	.704	.666	.623
8 I	.947	.916	.882	.845	.816	.792	.768	.745	.716	.671	.623	.563
8 S	.932	.898	.871	.842	.814	.784	.757	.728	.702	.664	.608	.561
9 I	.953	.936	.911	.895	.880	.864	.850	.835	.822	.804	.779	.742
9 S	.955	.936	.909	.889	.870	.856	.841	.829	.817	.799	.778	.749
10 I	.955	.926	.906	.889	.875	.857	.840	.823	.806	.781	.746	.708
10 S	.936	.900	.886	.865	.848	.835	.818	.807	.795	.774	.752	.724

I = Simulated Interstitial Lung Fluid; S = Simulated Surfactant Lung Fluid.

the data in Table 3 is

$$F = f_1 \cdot \exp[\lambda_1 t] + f_2 \cdot \exp[\lambda_2 t] + f_3 \cdot \exp[\lambda_3 t], \qquad (2)$$

with $f_1 + f_2 + f_3 = 1$. A simple transformation shows the equivalence between equations (1) and (2), i.e.

$$\lambda_i = \frac{-0.693}{t_{1/2}(i)}, \qquad \text{for } i = 1, 2 \text{ and } 3, \qquad (3)$$

where λ_i is defined as the rate of dissolution. This follows from

$$(1/2)f_i = f_i \cdot \exp[\lambda_i t_{1/2}(i)],$$

where f_i is defined as the initial total fraction. The parameters fit by the non-linear regression program were the three fractions of uranium (f_1, f_2 and f_3) and their associated dissolution half-times ($t_{1/2}(1)$, $t_{1/2}(2)$ and $t_{1/2}(3)$).

For each of the 20 sets of data given in Table 3, the program calculated estimates of the six parameters and their associated estimated asymptotic variances.

Table 4 gives the computer results for each of the 20 samples of the three fractions of uranium and their dissolution half-times. In addition, Table 4 gives the average values of the three fractions and half-times. Samples $2I$ and $3I$ appeared to be anomalies and were not included in the calculation of the average values for the six parameters.

The first question studied was whether there is a significant difference between the three fractions of uranium and their dissolution half-times for the two types of simulated lung fluids. This question can be answered by means of a t-test for each of the 10 samples and six parameters (three fractions and three half-times) which compares the two estimates of the parameters based on dissolving the sample in simulated interstitial

Table 4. Dissolution fractions and half-times of ten representative yellowcake samples obtained from one uranium milling operation dissolved in two simulated lung fluids

Sample	f_1	$t_{1/2}(1)$ (hours)	f_2	$t_{1/2}(2)$ (days)	f_3	$t_{1/2}(3)$ (days)
1_S^I	.195	3.39	.264	4.18	.541	52
	.257	4.17	.171	3.09	.572	40
2_S^I	.162	3.78	.428	8.19	.411	121
	.192	4.08	.162	3.86	.645	54
3_S^I	.201	3.29	.415	12.38	.384	534
	.159	3.38	.186	4.57	.655	94
4_S^I	.193	3.38	.288	6.68	.519	509
	.169	3.35	.230	5.33	.601	187
5_S^I	.136	2.75	.167	4.61	.697	362
	.142	2.85	.176	5.81	.682	806
6_S^I	.105	4.13	.157	4.88	.737	324
	.119	4.15	.151	4.24	.730	264
7_S^I	.148	3.19	.131	4.01	.721	126
	.174	3.71	.142	4.23	.684	193
8_S^I	.177	3.73	.187	5.00	.635	144
	.176	3.18	.200	4.27	.624	165
9_S^I	.117	3.13	.069	3.06	.814	200
	.131	3.54	.073	3.83	.796	303
10_S^I	.120	2.95	.115	4.31	.765	234
	.145	2.56	.081	3.36	.774	275
Average Value*	.160	3.45	.165	4.39	.675	224

I = Simulated Interstitial Lung Fluid; S = Simulated Surfactant Lung Fluid

*Samples 2S and 3S were used to represent Samples 2 and 3.

and simulated surfactant lung fluids. The 60 t-statistics are given in Table 5 and were computed by the following formula:

$$t = \frac{I - S}{(S_I^2 + S_S^2)^{1/2}},$$

where I and S are the estimates of the parameter with the sample dissolved in simulated interstitial and simulated surfactant lung fluids and S_I^2 and S_S^2 are estimated asymptotic variances of these two estimates (computed by the non-linear regression program). Each parameter estimate and its associated estimated variance has 7 degrees of freedom (df); this is computed by the following formula:

The sixth parameter, f_3, was constrained as stated after equation (1), i.e. $f_3 = 1 - (f_1 + f_2)$.

Preliminary F-tests with $\alpha = 0.01$ were performed to test the assumption for the t-tests of equality of the variances, $\sigma_I^2 = \sigma_S^2$. The F-statistic was significant in 12 of the 60 cases, all of which involved either the anomalous Samples 2 and 3 or the $t_{1/2}(3)$ estimates which have large variances. The 99% critical value for these 12 t-tests is $t_{0.01}(7 \text{ df}) = \pm 3.499$, since the degrees of freedom should not be added together. For the other 48 t-tests, the 99% critical value is $t_{0.01}(14 \text{ df}) = \pm 2.977$. The significance level, α, was chosen to be 1% since 3 of the 60 t-tests would be expected to be significant by chance

(number of data points) − (number of free parameters estimated) = 12 − 5 = 7.

Table 5. t-statistics for comparison of uranium fractions and dissolution half-times between simulated lung fluid pairs

Sample Pair	f_1	$t_{1/2}^{(1)}$	f_2	$t_{1/2}^{(2)}$	f_3	$t_{1/2}^{(3)}$
1	-1.280	-.620	.628	.285	-.180	.325
2	-.906	-.243	.433	.432	-.372	.083
3	1.473	-.087	.148	.235	-.173	.013
4	.807	.026	.177	.166	-.235	.049
5	-.351	-.125	-.066	-.240	.105	-.084
6	-.501	-.014	.057	.128	.052	.073
7	-1.020	-.547	-.118	-.051	.338	-.249
8	.041	.545	-.076	.135	.063	-.071
9	-.802	-.512	-.102	-.205	.356	-.360
10	-2.356	.837	.861	.407	-.202	-.209

alone if 95% critical values ($\alpha = 0.05$) had been used. None of the t-statistics exceeded its critical value. Therefore, the conclusion is that there is no significant difference between the two types of simulated lung fluids with respect to the fractions of uranium undissolved and their dissolution half-times.

The second question studied was whether there is a significant difference among the 10 samples with respect to the three fractions of uranium and their respective dissolution half-times. This question can be answered by performing all pairwise comparisons among the 10 samples using a multiple comparison procedure for each of the six parameters. The average of the two parameter estimates (I and S) and the average of the two estimated variances (S_I^2 and S_S^2) are used, except for Samples 2 and 3, since the t-tests showed no significant differences in the parameter estimates between the two lung fluids. For Samples 2 and 3, the results in Table 4 for $2S$ and $3S$ were used to represent these samples instead of the average of the I and S results. The pairwise comparisons among the 10 estimates can be performed by the Student–Newman–Keuls test if the 10 variances can be assumed to be equal (see a statistical text, e.g. Sn67), or by the Brown–Forsythe test if the 10 variances are not equal (Br74). Therefore, a preliminary test on the equality of the 10 variances, Bartlett's test (e.g. see Sn67), was made. As a result, the Brown–Forsythe test was performed on all six parameters, since Bartlett's test indicated large differences among the 10 variances. These tests were performed at the 5% significance level. No pair of estimates from among the 10 samples and six parameters was judged significantly different. Therefore, the conclusion is that there are no significant differences among the 10 estimates for the three uranium fractions and three dissolution half-times and there are no significant differences in the dissolution characteristics among the 10 samples of yellowcake. It should be noted that equation (1) was fitted using data from a period of 26.5 days and that the average value for the half-time of the third fraction is 224 days. Seemingly large differences in the estimates of the half-times of the third fraction appear in Table 4. No statistically significant differences among the 20 dissolution half-times were identified, because of the large variances associated with the point estimates of the dissolution half-time of the third fraction.

Since there were no significant differences between the interstitial and surfactant fluids and no significant differences among the 10 samples of yellowcake, the 10 estimates of each of the six parameters can be used to

calculate point estimates and confidence intervals for the three fractions of uranium and their dissolution half-times. The confidence intervals for the parameters were computed using the mean of the 10 values and the standard deviation of the mean. The point estimates can be used in equation (1) to obtain a prediction equation for the fraction of uranium remaining undissolved at a particular time for this milling operation. The confidence intervals indicate the precision with which the parameters are estimated. See Table 6 for the point estimates and 95% confidence intervals for the three uranium fractions and three dissolution half-times.

When data from this investigation are compared to previous investigations of yellowcake (Ka78; Ei80), it is strongly suggested that significant differences do indeed exist among yellowcake products obtained from industry. The product utilized in this investigation was obtained from the mill whose product was reported by previous investigations to be the most insoluble among the four samples. Although the scope of this investigation did not include analysis of yellowcake from the other three mills previously investigated, information regarding thermal history of each product was obtained. It was determined that the thermal history of the samples investigated in this study was significantly higher in temperature than the three other yellowcake products previously investigated. The three other uranium milling samples investigated by Kalkwarf (Ka78) and Eidson and Mewhinney (Ei80) were representative of yellowcake obtained from mills which utilize low-temperature drying of yellowcake, at temperatures less than 400°C (782°F). Samples obtained from the single source utilized in this investigation and the remaining sample investigated by others (Ka78; Ei80) are representative of uranium mill samples obtained from mills that utilize high-temperature drying which actually calcines the yellowcake at temperatures generally much greater than 400°C. It appears that a single generalized solubility classification cannot be made for all yellowcake obtained from industry. More appropriately, it appears that industrial yellowcake could be classified by two solubility equations, one representative of high-temperature calcined yellowcake and the other for low-temperature dried yellowcake. These assertions regarding the significance of thermal history appear to be supported by earlier research (St66; Co74).

Other factors which may have also contributed to differences among the yellowcake samples from the four mills include specific ore body and its characteristics and the chemical extraction techniques utilized. These factors were not included in the scope of this work.

CONCLUSIONS

Yellowcake dissolution in simulated interstitial and simulated surfactant lung fluids is not significantly different, based on data over the first 26.5 days. The average dissolution rates of yellowcake examined in this investigation can be described as the sum of three exponential terms. The solubility

Table 6. Estimates and confidence intervals for uranium fractions
and dissolution half-times

Parameter	Point Estimate	95% Confidence Interval
f_1	16.0%	13.5% to 18.5%
$t_{1/2}(1)$	3.45 hours	3.12 hours to 3.78 hours
f_2	16.5%	12.6% to 20.4%
$t_{1/2}(2)$	4.39 days	3.83 days to 4.95 days
f_3	67.5%	61.6% to 73.3%
$t_{1/2}(3)$	224 days	107.8 days to 340.4 days

classification of yellowcake obtained from one milling operation can be approximated as 32.5% D 67.5% Y, although two distinct fractions can be identified within the class D fraction. Chemical variations, as well as variations in particle size and surface area of particles, exist among yellowcake samples; however, density appears to be relatively homogeneous among product from a single mill. This investigation of samples from a single source indicates no significant variability in dissolution fractions and half-times. These fractions and half-times, when compared with dissolution studies of yellowcake from other mills, indicate that the dissolution properties of yellowcake may, indeed, be unique for a specific site or process method.

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