From: John Schmuck [John_Schmuck@Cameco.com]
Sent: Tuesday, September 13, 2011 3:58 PM
To: Burrows, Ronald
Subject: RE: Information in Response to August 8, 2011 Discussion

Yes sir, you may make it public on both dockets.

.john

-----Original Message-----
From: Burrows, Ronald [mailto:Ronald.Burrows@nrc.gov]
Sent: Tuesday, September 13, 2011 1:57 PM
To: John Schmuck
Cc: Mandeville, Douglas
Subject: RE: Information in Response to August 8, 2011 Discussion

Thank you, John.

There is a "confidential" marking memo.

Can I make this publicly available on both dockets?

Thank you.

Ron

From: John Schmuck [mailto:John_Schmuck@Cameco.com]
Sent: Tuesday, September 13, 2011 3:06 PM
To: Burrows, Ronald
Subject: Information in Response to August 8, 2011 Discussion

Ron - Attached please find technical clarifications related to the solubility study. Because this information pertains to both Crow Butte and Smith Ranch entry into each site's administrative record may be appropriate.

If you have additional questions, please do not hesitate to call.

Thanks. .john

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From: John Schmuck [John_Schmuck@Cameco.com]
Sent: Tuesday, September 13, 2011 3:06 PM
To: Burrows, Ronald
Subject: Information in Response to August 8, 2011 Discussion
Attachments: Lung Study Clarification Response 8-11.pdf

Ron - Attached please find technical clarifications related to the solubility study. Because this information pertains to both Crow Butte and Smith Ranch entry into each site's administrative record may be appropriate.

If you have additional questions, please do not hesitate to call.

Thanks. .john

Interoffice Memo Cameco

Date: September 9, 2011 [redacted]

To: Mike Murchie File: 200183

From: Gioulchen Tairova

Re: **Solubility of Smith Ranch-Highland and Crow Butte Operation Uranium Ore Concentrate Samples in Simulated Lung Fluid**

EXECUTIVE SUMMARY

The US NRC representative requested additional information on the composition and particle size distribution of Smith Ranch-Highland (SRH) and Crow Butte operation (CBO) uranium ore concentrate (UOC) samples used in the 2009 studies of dissolution in simulated lung fluid. The available relevant information was summarized in this report. Results of XRD analyses indicated that the major crystalline uranium compounds in SRH UOC samples were metaschoepite and uranium trioxide hydrate, and in CBO UOC samples, metaschoepite and metastudtite. Small quantities of unidentified amorphous third phase might also be present in the samples. The mean particle diameter of the <20 μm fraction for the SRH sample determined by SEM image analysis was 5 μm with a standard deviation of 8.8 μm. The particle size distribution analysis using sedimentation method indicated that the median particle diameter of the <20 μm fraction for CBO and SRH samples was 10.6 μm and 11.9 μm, respectively.

1.0 INTRODUCTION

The studies of the dissolution of SRH and CBO UOC samples in simulated lung fluid (SLF) were carried out in 2009 and the results were summarized in a report [1].

During a teleconference held on August 8, 2011 among personnel of Cameco Resources Inc., US NRC and Cameco technology and innovation-research centre (CTI-RC) to discuss the results from the above report, the US NRC representative requested further information on the following questions:

1) What was the composition of the samples used in the experiments?
2) Why the <20 μm fraction was used in SLF studies and what was the particle size distribution in the studied samples?
3) Why the two- or three-term equations were used for assignment of a solubility type?

This report provides answers to the above questions based on relevant information on hand.

2.0 RESULTS AND DISCUSSIONS

2.1 Composition of Samples – XRD Results

2.1.1 Smith Ranch-Highland Samples

Three SRH UOC samples were analyzed using X-ray diffractometry (XRD). The results of the XRD analyses indicated the presence of crystalline metaschoepite, $UO_3 \cdot 2H_2O$, and uranium trioxide hydrate, $UO_3 \cdot 0.8H_2O$, in all three samples. Small quantities of unidentified amorphous phase might also be present. The X-ray diffractograms for the SRH samples are shown in Figures 1-3 [2].

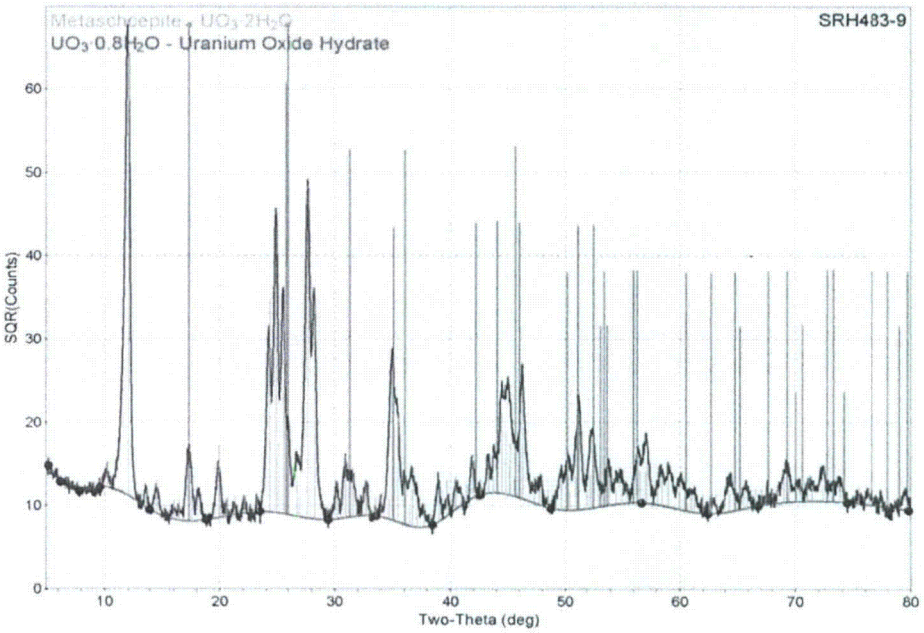

Figure 1. The X-ray diffractogram for the SRH sample No. SRH 483-9

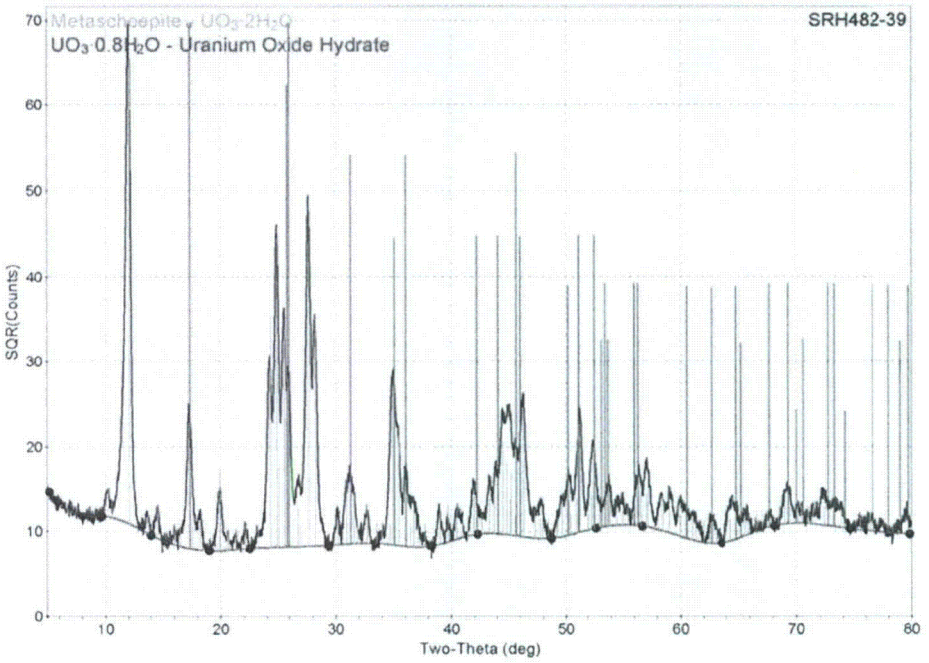

Figure 2. The X-ray diffractogram for the SRH sample No. SRH 482-39

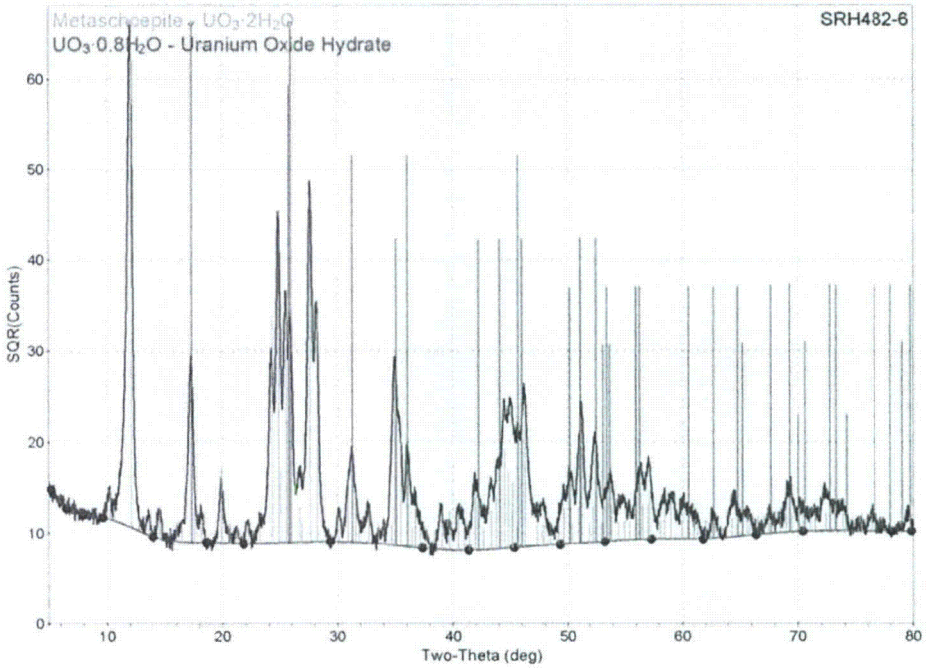

Figure 3. The X-ray diffractogram for the SRH sample No. SRH 482-6

The quantities of an amorphous third phase, which is apparent from the elevated baselines in Figures 1-3, is smallest in the sample No. SRH 482-6.

2.1.2 Crow Butte Operation Samples

Results of XRD analyses of CBO UOC samples indicated that crystalline phases of metaschoepite, $UO_3 \cdot 2H_2O$, and metastudtite, $UO_4 \cdot 2H_2O$, were found in all samples [2]. The X-ray diffractograms for the CBO samples are shown in Figures 4 - 6.

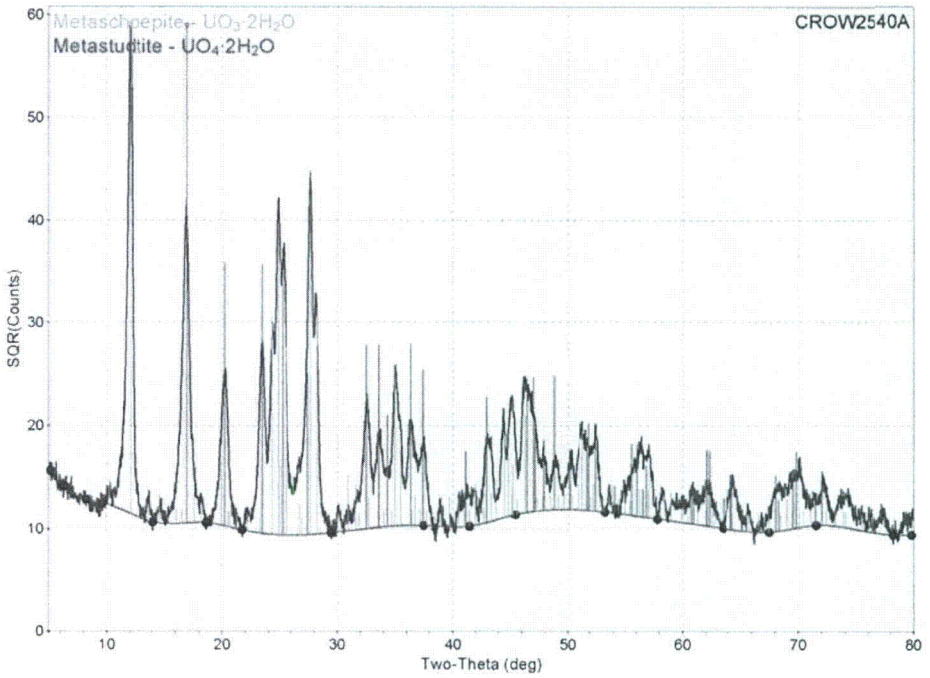

Figure 4. The X-ray diffractogram for the CBO sample No. Crow 2540A

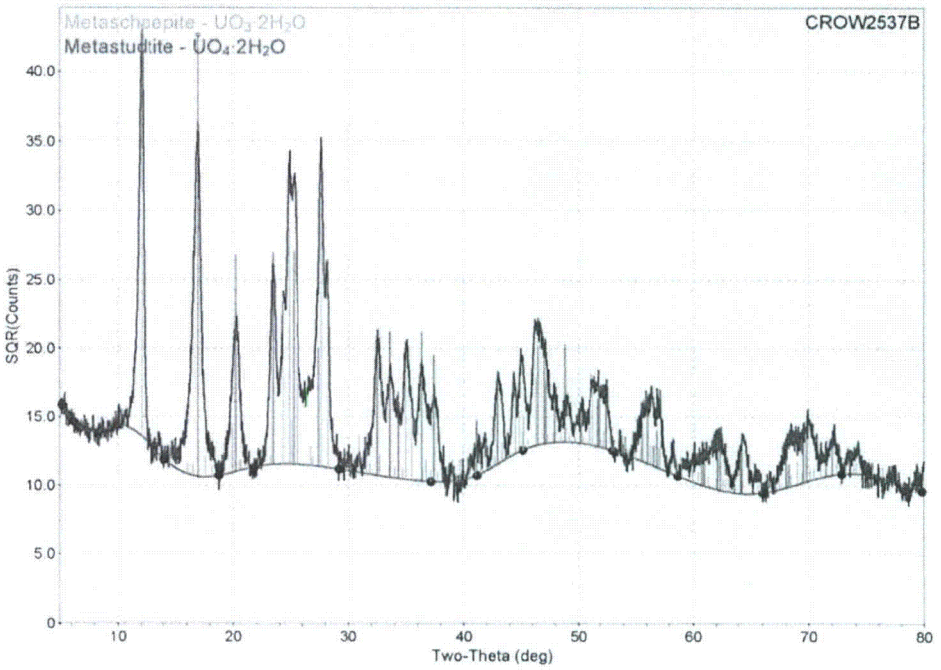

Figure 5. The X-ray diffractogram for the CBO sample No. Crow 2537B

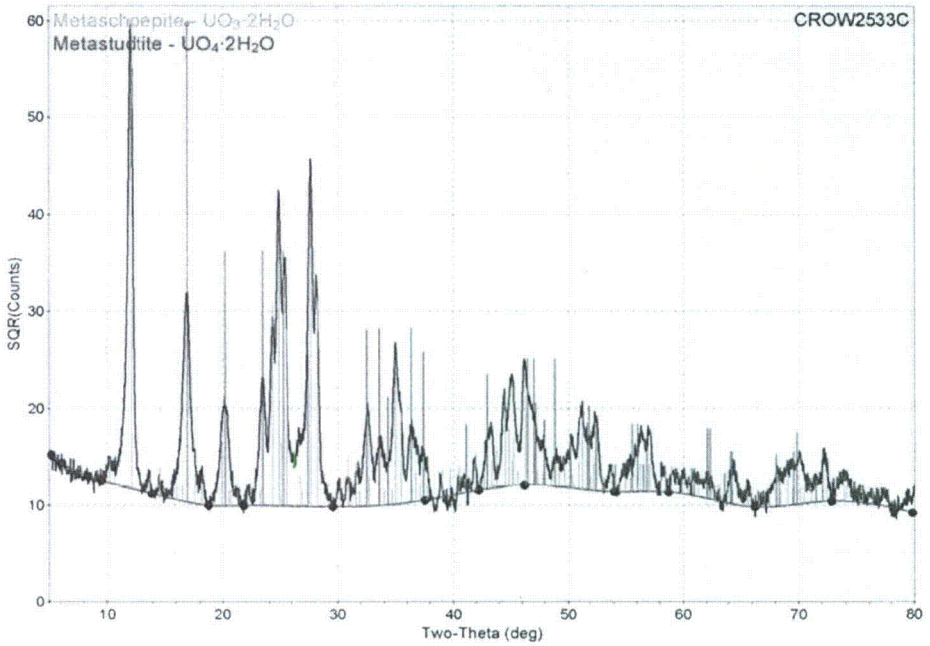

Figure 6. The X-ray diffractogram for the CBO sample No. Crow 2533C

The elevated baselines in Figures 4-6 also indicated the potential presence of various quantities of unidentified amorphous third phase.

2.2 Particle Size of Samples

In the literature, fractions with a wide range of particle size distribution were used for simulated lung fluid dissolution studies. The fractions of particles applied varied from less than 1 μm to less than 25 μm [3,4].

The latest guidance from ICRP Publication 66 (ICRP 1994) and Publication 78 (ICRP 1998), cited in a 2006 NIOSH study, suggested the 5-μm AMAD[1] as the default median particle size [5].

For all simulated lung fluid dissolution experiments conducted at the research centre, samples were sieved to obtain the fraction less than 20 μm to simulate airborne samples.

SEM Morphology Studies

The shape and diameter of particles in the as-received SRH and CBO UOC samples were observed with a scanning electron microscope (SEM). Most of the particles were spherical in shape. Overview images of four areas of a typical Crow Butte UOC sample at 1000X magnification are shown in Figure 7, a through d [6]. The sample consisted of particles/agglomerates ranging in size from sub-micron to approximately 100 μm.

[1] Activity Median Aerodynamic Diameter (AMAD) is the diameter in an aerodynamic particle size distribution for which the total activity above and below this size are equal. A log-normal distribution of particle sizes is assumed.

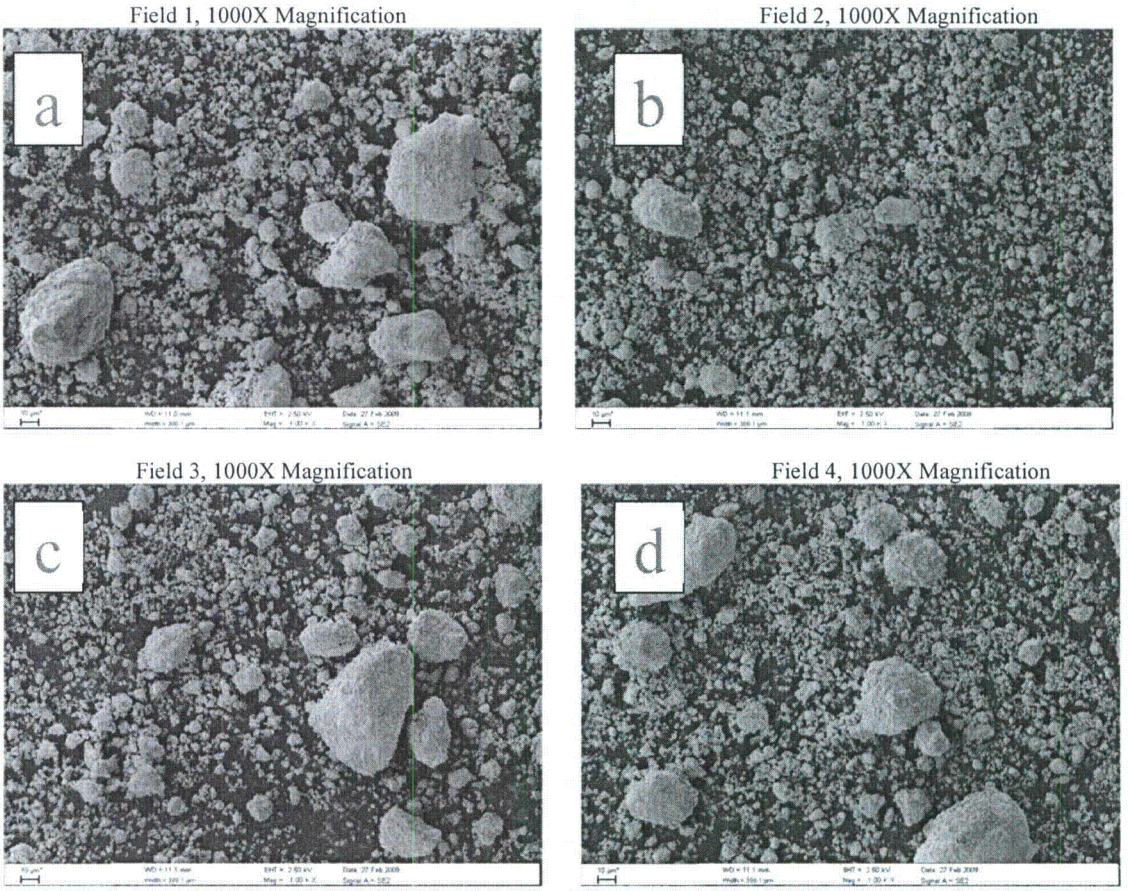

Figure 7. Scanning electron micrographs of CROW-2537-A

To determine the particle size using the SEM technique, secondary SEM electron images were collected in ten fields across the sample surface at 500X magnification. Image analysis software was used to characterize the particles. Unfortunately, due to a high population of fine particles (<5 μm) in the images, the software was not able to generate a particle size histogram. However, the SEM images provided some information on actual sizes of particles.

Overview SEM images of two areas of a typical Crow Butte UOC sample of a <20 μm fraction recorded at 500 X magnification are shown in Figure 8. It can be seen that a large population of particles are finer than 5 μm.

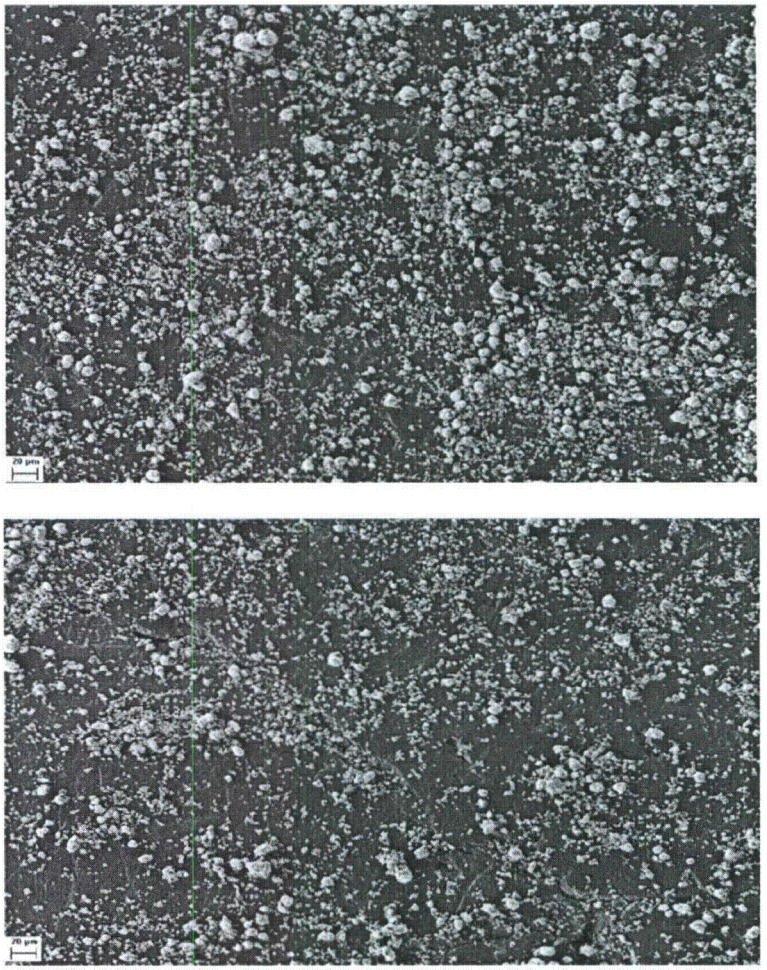

Figure 8. **SEM images of a CBO sample No. CROW 2537C <20 μm fraction at 500X magnification**

The SEM images of a typical as-received SRH UOC sample are shown in Figure 9, a through d [7]. The sample consisted of particles/agglomerates ranging in size from sub-micron to approximately 50 μm.

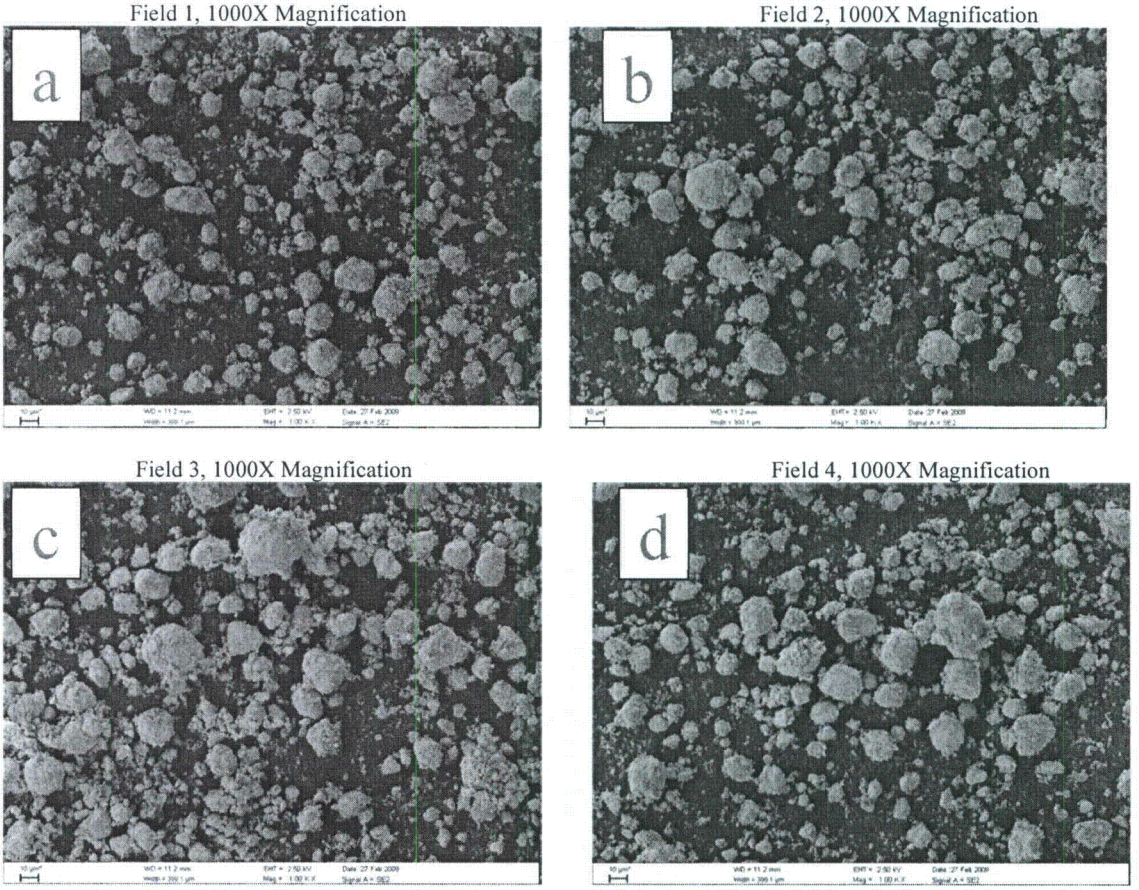

Figure 9. SEM images of as-received SRH sample No. SRH 482-13 at 1000X magnification

To determine the particle size using the SEM technique, secondary SEM electron images were collected in ten fields across the sample surface at 500X magnification. Image analysis software was used to characterize the particles. The feature used to characterize particle size is referred to as "length" in the software. This is defined as the longest axis in any direction through a given particle.

A summary of the data collected from the SEM analysis is presented in Table 1. The analysis indicated that the mean particle size was 5 μm with a standard deviation of 8.8 μm.

A histogram showing the particle size distribution in the SRH482-29 sample of a <20 μm fraction is shown in Figure 10.

Table 1. Particle Size Data

Analysis Statistics	Particle Diameter (μm)
Mean Particle Size	5.0
Standard Deviation	8.8
Minimum	0.59
Maximum	117
Number of Particles Measured	2225

There is some degree of uncertainty to the accuracy of these results. The minimum particle size detectable at 500X magnification was 0.59 μm. Therefore, any particles smaller than this were not counted.

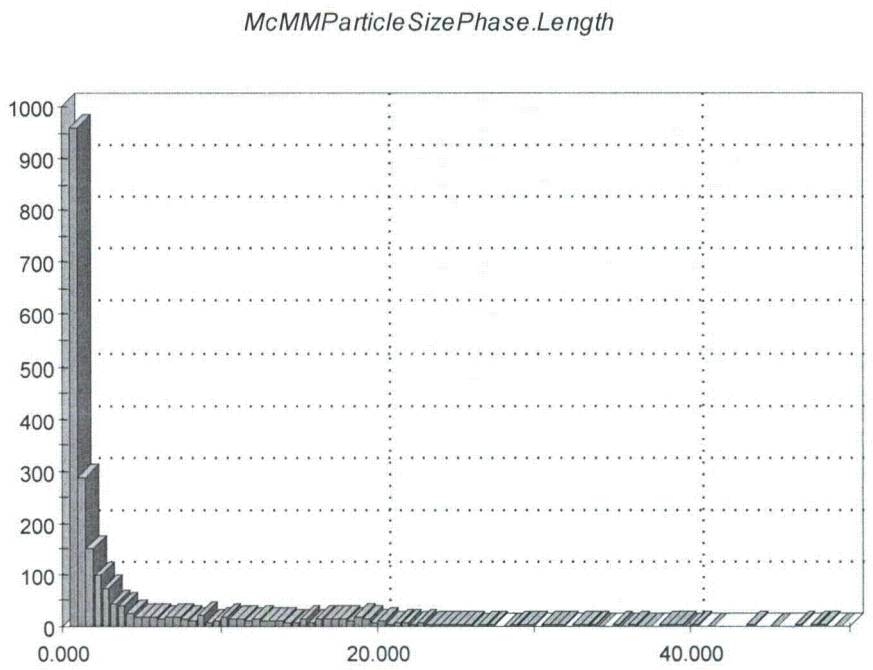

Figure 10. Particle size distribution from SEM image analysis for sample SRH 482-29

Overview SEM images of two areas of a typical SRH sample of a <20 μm fraction recorded at 500X magnification are shown in Figure 11 [8]. It can also be seen that a large population of particles are at the 5 μm size or finer.

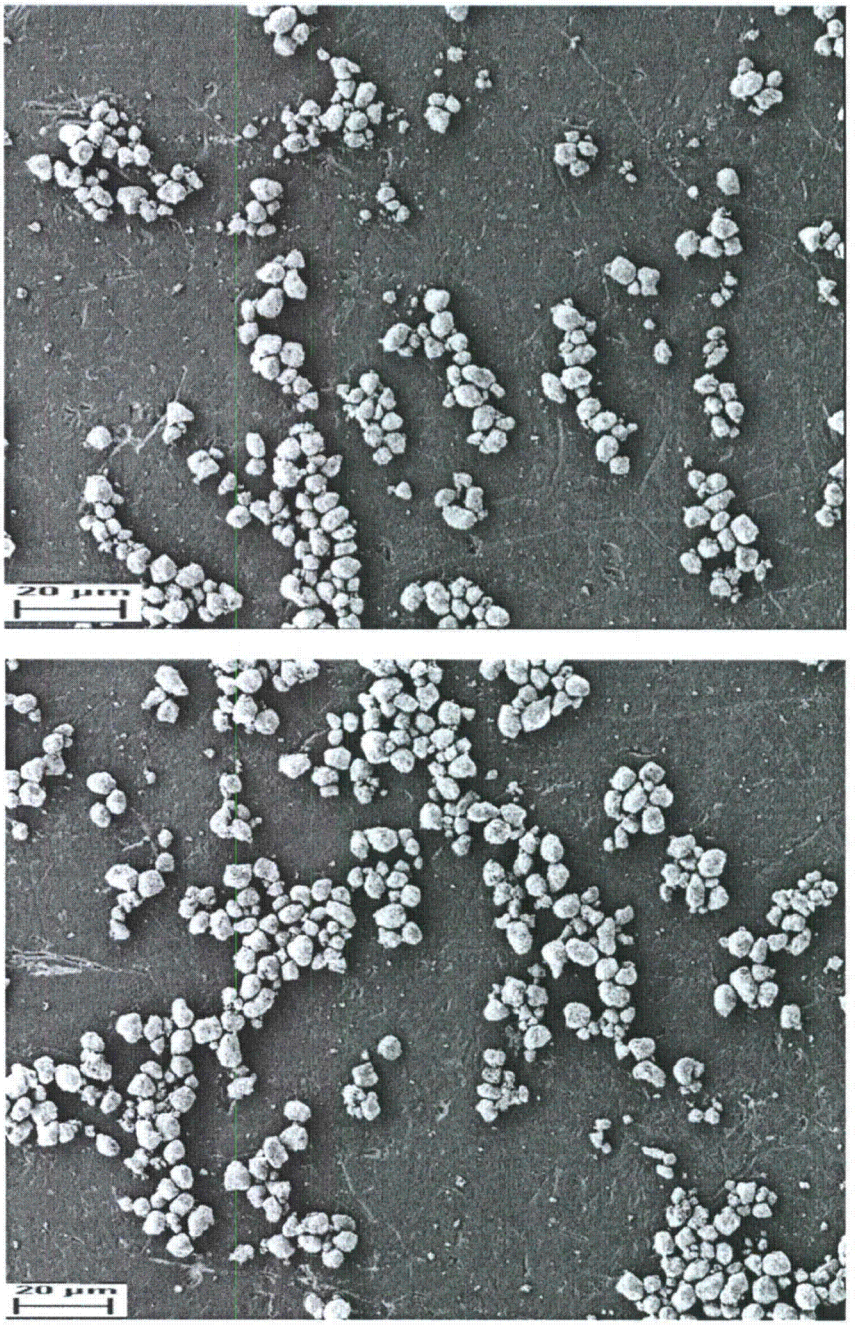

Figure 11. **SEM images of SRH sample No. SRH 482-29, <20 μm fraction, at 500X magnification**

X-Ray Sedimentation Analysis

The particle size distribution study for CBO and SRH samples of a <20 μm fraction, using the X-ray sedimentation technique, indicated that a median particle diameter was 10.6 μm and 11.9 μm, respectively. The particle size distribution was shifted to a higher median diameter from the mean diameter, determined by SEM image analysis, due to possible partial agglomeration (not complete dispersion) of particles.

Particle size distributions for the <20 μm fraction of CBO and SRH samples are shown in Figure 12.

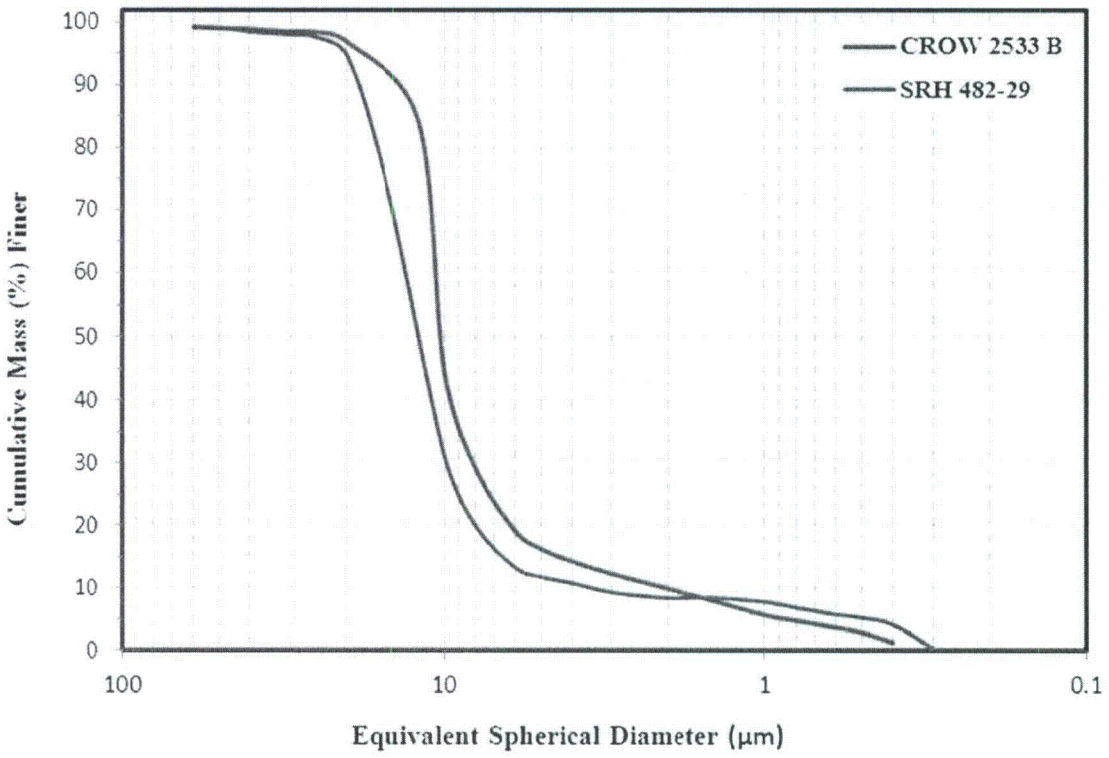

Figure 12. Particle size distributions of <20 μm fraction of CBO and SRH UOC samples CROW2533B and SRH 482-29, respectively

2.3 Solubility in Simulated Lung Fluid

Smith Ranch-Highland Samples

Uranium dissolution kinetics, related parameters and absorption Types for the three SRH samples, calculated based on the ICRP 66 and ICRP 71 criteria, were reported earlier [1].

The kinetics data from the tests performed on SRH and CBO UOC samples were fitted using a three or two-exponential equation to classify the material as DWY type. This allowed estimating the dissolution times and the fraction of material assigned to each type. The number of terms used in the equation was based on statistical analysis. The equation with the lowest error was

selected for each sample. For two SRH samples, the equation with the lowest error was three-term equation:

$$\frac{M}{M_0} = f_1 \exp(-0.693\frac{t}{T_1}) + f_2 \exp(-0.693\frac{t}{T_2}) + f_3 \exp(-0.693\frac{t}{T_3}) \quad (1)$$

Where:

M - mass of undissolved uranium at time t

M_0 - initial mass of uranium

t - elapsed time

f_1 - fraction of total U with corresponding dissolution half-time T_1

f_2 - fraction of total U with corresponding dissolution half-time T_2

f_3 - fraction of total U with corresponding dissolution half-time T_3

$f_1 + f_2 + f_3 = 100\%$

For one SRH sample, a two-term equation had slightly better fit. The two-term equation applied was:

$$\frac{M}{M_0} = f_1 \exp(-0.693\frac{t}{T_1}) + f_2 \exp(-0.693\frac{t}{T_2}) \quad (2)$$

A program was developed at the CTI-RC for calculation of dissolution parameters using non-linear regression analysis [9].

The standard deviations of parameters were calculated from the inverse Hessian matrix using the mean square error (MSE) calculated as the square root of F/(N-v), where **N** is the number of data points and **v** is the number of parameters used in the model (**N-v** is the number of degrees of freedom).

The program automatically tests several kinetics models applied to the same set of experimental data. For each model, the minimization is repeated 200 to 500 times (this number is specified by the user). Then, the program selects the model that has the smallest MSE, which is considered to be the best approximation to the experimental data. Some models with a large number of adjustable parameters that show lower F are not the best description for the experiment because they have higher MSE (lower denominator).

In some cases, when calculated values of uncertainties are high, an additional sampling during first day and (or) after last day of extraction is required in order to obtain lower uncertainties.
The value of the mean square deviation (MSD) characterizes an average error for the curve fitting, i.e., the difference between the experimental values of uranium extraction, $(1 - M/M_0)*100\%$, and the theoretical value.

The use of equations (1) or (2) allows one to determine the rapid and slow fractions and their corresponding half-times. Parameters obtained can be used to calculate the dose intake according to ICRP Publication 66.

The uranium dissolution parameters for three randomly selected samples, calculated using the two- and three-exponential models, as well as the assignment to DWY solubility Types, based on the ICRP 30 classification, used by the US NRC, are shown in Table 2.

The presence of different quantities of an amorphous third phase could explain the better fit for two- or three-term exponential equation. As was mentioned earlier, for the sample SRH 482-6, the two-exponential equation had slightly better fit (MSD=4.0), than the three exponential (MSD=4.1).

Table 2. Uranium dissolution parameters for SRH samples

Sample No. [a]	f_1 (%)	T_1 (d)	f_2 (%)	T_2 (d)	f_3 [a] (%)	T_3 (d)	MSD [b] (%)	Type D	Type W
SRH 482-6	84.5	1.6	15.5	18.2			3.8	84.5	15.5
SRH 482-39	53.4	0.04	33.3	1.2	13.3	60	1.7	88.6	13.3
SRH 483-9	48.5	0.02	34.7	0.34	16.8	2.7	0.4	100	

Note: [a] The results of the analysis indicated only traces of residual uranium on a filter after 100 d of extraction
[b] The two- or three- exponential models were applied for calculations

The mass balance, based on the residual uranium, measured using X-ray fluorescence method, and the total amount of uranium calculated from the uranium extracted in each sample during 100 days of extraction, is shown in Table 3. The results of the analysis indicated only traces of residual uranium on a filter after 100 days of extraction.

Table 3. Comparison of calculated and measured residual uranium quantities in SRH samples

Description	Sample No.		
	SRH 482-6	SRH 482-39	SRH 483-9
U total in 50 mg sample, (%)	79.4	78.7	79.4
U in a sample initial (mg)	39.68	39.33	39.7
U total extracted calculated from combined extracted U in each sample (mg)	38.6	37	39.6
U residual on a filter after the test completion, calculated (mg)	1.1	2.3	0.1
U residual on a filter, XRF analyses after the test completion (mg)	0.4	0.5	0.1
Difference (U residual on a filter calculated and measured)	0.7	1.8	0.0
Standard error (%)	1.7	4.7	0.0

Crow Butte Operation Samples

Uranium dissolution kinetics, dissolution parameters and absorption types for the CBO samples, based on the ICRP 66 and ICRP 71 criteria were reported earlier [1]. The uranium dissolution parameters for three randomly selected samples, calculated using the two-exponential and three-exponential models, as well as the assignment to DWY solubility Types based on the ICRP 30 classification, used by the US NRC, are shown in Table 4.

Table 4. Dissolution parameters and absorption types for uranium in CBO concentrate samples

Sample No.	f_1 (%)	T_1 (d)	f_2 (%)	T_2 (d)	f_3 [a] (%)	T_3 (d)	MSD [b] (%)	Type D	Type W
CROW 2540-A	45.7	0.42	47.1	5.03	7.2	>100	1.6	92.8	7.2
CROW 2537-B	43.6	0.3	50.6	4.89	5.9	>100	2	94.1	5.9
CROW 2533-C	90.0	1.5	10.0	76			2.9	90.0	10.0

Note: [a] The results of the analysis indicated only traces of residual uranium on a filter after 100 d of extraction
[b] The two- or three- exponential models were applied for calculations

The mass balance, based on the residual uranium, measured using X-ray fluorescence method, and the total amount of uranium calculated from the uranium extracted in each sample during 100 days of extraction, is shown in Table 5. The results of the analysis indicated only traces of residual uranium on a filter after 100 d of extraction, which were significantly lower than the calculated f_3 fraction, shown in Table 5. Based on the results of the analysis of residual uranium, within experimental errors, the calculated f_3 fraction could be assigned to Type W, not Y.

Table 5. Comparison of calculated and measured residual uranium quantities in CBO samples

Description	Sample No.		
	CROW 2540-A	CROW 2537-B	CROW 2533-C
U total in 50 mg sample, (%)	73.12	73.44	74.24
U in a sample initial (mg)	36.56	36.72	37.12
U total extracted calculated from combined extracted U in each sample (mg)	33.9	34.43	35.04
U residual on a filter after the test completion, calculated (mg)	2.66	2.29	2.08
U residual on a filter, XRF analyses after the test completion (mg)	0.3	0.8	0.7
Difference (U residual on a filter calculated and measured)	2.36	1.49	1.38
Standard error (%)	6	4	4

3.0 CONCLUSIONS

The results of the XRD analyses indicated the presence of crystalline phases of metaschoepite and uranium trioxide hydrate in SRH samples and metaschoepite and metastudtite in CBO samples. Small quantities of unidentified amorphous third phase might also be present in the samples. The presence of different quantities of a third amorphous phase could explain the better fit for two- or three-term exponential equation.

The mean particle diameter of the <20 μm fraction of the SRH sample determined by SEM image analysis was 5 μm with a standard deviation of 8.8 μm. The particle size distribution analysis using sedimentation method indicated that the median particle diameter of the <20 μm fraction for CBO and SRH samples was 10.6 μm and 11.9 μm, respectively.

4.0 REFERENCES

[1] G. Tairova, "Solubility of Radionuclides in Simulated Lung Fluid. Part 3: ISR and RLO Samples", Cameco Technology and Innovation-Research Centre, Project 200183, Report No. RC-09-05, October 2009.

[2] L.B. Johnson, "X-Ray Diffraction Tests of SRH483-9, SRH482-39, SRH482-6, CROW2540-A, CROW2537-B and CROW2533-C", Cameco Technology and Innovation-Research Centre, interoffice memorandum, June 30, 2010.

[3] V. Chazel, P. Houpert and E. Ansoborlo, "Effect of U_3O_8 Specific Surface Area on In-Vitro Dissolution, Biokinetics and dose Coefficients", Radiation Protection Dosimetry, 1998, Vol. 79, No. 1-4, 39-42.

[4] S. Sdraulig, R. Franich, R.A. Tinker, S. Solomoon, R. O'Brien and P.N. Johnson, "In Vitro Dissolution Studies of Uranium Bearing Material in Simulated lung Fluid", J. of Environmental Radioactivity, 2008, Vol. 99, 527-538.

[5] "Dose Reconstruction Project for NIOSH", Y-12 National Security Complex–Occupational Internal Dose, Report No. ORAUT-TKBS-0014-5, Effective Date: 01/12/2006.

[6] W.C. Hubers, "SEM Morphological Examination of CROW-2537-A", Cameco Technology and Innovation-Research Centre interoffice memorandum, February 27, 2009.

[7] W.C. Hubers, "SEM Morphological Examination of SRH 482-13", Cameco Technology and Innovation-Research Centre interoffice memorandum, February 27, 2009.

[8] W.C. Hubers, "SEM Particle Size Analysis of Sample SRH 482-29", Cameco Technology and Innovation-Research Centre interoffice memorandum, August 31, 2011.

[9] G. Tairova and M. Ioffe, "Solubility of Radionuclides in Simulated Lung Fluid", Cameco Technology and Innovation-Research Centre, Project 200183, Report Number TD 02-14, September 2002.