Cameco

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September 17, 2010

Mr. Ronald A. Burrows
NRC, Program Manager
Decommissioning & Uranium Recovery Licensing
Div. of Waste Management & Environmental Protection
Federal/State Materials & Environ. Mang. Programs
Mail Stop T8-F5
Washington DC 20555-0001

Subject: Response to letter dated June 30, 2010, Open Issues, License Amendment, Crow Butte Resources, Inc., Crawford Nebraska, License SUA-1534 (TAC J00555)

Dear Mr. Burrows:

By letter dated June 30, 2010, the U.S. Nuclear Regulatory Commission identified open issues associated with its preparation of a draft of the Safety Evaluation Report and requested additional information. Attached please find the Crow Butte Resources responses to the open issues and associated additional information. On this occasion, Crow Butte Resources has targeted October 1, 2010 for separate transmittal of the corresponding replacement pages.

If you or your staff has any questions, please contact John Schmuck at (307) 316-7587.

Sincerely,

Tom Young,
Vice-President, Operations

Attachments: CBR Responses

CC: Jim Stokey, no attach Larry Teahon, w/attach John Schmuck, w/attach
Josh Leftwich w/attach CR file w/attach

Responses to NRC Open Issues, License Renewal Amendment Request, Crow Butte Resources, Inc., Crawford Nebraska, License SUA-1534 (TAC J00555), dated June 30, 2010

I. Open Issues

A. Section 4.1.1, Tank and Process Vessel Ventilation Systems – In Section 4.1.1 of the Technical Report (TR), the applicant describes the negative pressure maintained on the vacuum dryer and during packaging operations. Please describe the location of the exhaust from each of these vacuum sources and the area receiving the exhaust.

***Response**: The limited, intermittent and variable exhaust flow from the vacuum pump is returned to the drying and packaging room. As described in Section 5.8.3.1, the air in the drying and packaging room circulates through a HEPA filter while the room is maintained as an Airborne Radioactivity Area. Air sampling occurs to verify that the airborne radioactivity concentrations are within acceptable limits before the room is released from the Airborne Radioactivity Area status.*

B. Section 5.6.1.3, Training Content – In Section 5.6.1.3 of the TR, Crow Butte Resources, Inc. (CBR) discusses its training program content for its employees. However, there is no specific discussion of the instructions regarding risks from occupational radiation exposure and prenatal radiation exposure. Consistent with NUREG-1569, Acceptance Criteria 5.5.3(2) and 5.5.3(3), please address how these instructions are incorporated into the applicant's training program.

***Response**: RGs 8.13 and 8.29 have been specifically identified in the text of Section 5.6.1.3.*

C. Section 5.8.3.1, Airborne Uranium Particulate Monitoring

1. In CBR's May 12, 2009 letter (ML091470119) responding to NRC staff's Request for Additional Information (ML083660060), it did not provide sufficient information to justify the use of 100 percent Class D yellowcake. Please provide technical justification for assuming 100 percent Class D inhalation class for the yellowcake produced at the Crow Butte Project. In addition, if the proposed inhalation class changes, provide an appropriate revised lower limit of detection for measuring uranium in air.

***Response**: As a standard, the NRC has traditionally used Class D solubility for natural uranium as the inhalation class when approving ISL license applications and renewals and when performing compliance inspections. That approach is consistent with Reg. Guide 8.30, where NRC stated:*

> *"Yellowcake dried at low temperature, which is predominantly composed of ammonium diuranate, or in the new processes uranyl peroxide, both are more soluble in body fluids than yellowcake dried at higher temperature; and a relatively large fraction is rapidly transferred to kidney tissues (Refs. 9 to 11). If the intake of such yellowcake is controlled to protect the kidney from the chemical toxicity of uranium, radiological protection criteria for natural uranium will also be satisfied. For purposes of compliance with 10 CFR Part 20, yellowcake undried or dried at low temperature should be classified as soluble.*
>
> *Yellowcake dried at high temperature is a mixture of compounds that contains a major portion of more insoluble uranium oxides. Radiation dose to the lung and other organs is the limiting consideration rather than chemical toxicity; this is primarily due to the large insoluble component. For compliance purposes, yellowcake dried at 400°C (752°F) and above should be classified as insoluble (Refs. 12 and 13)."*

Cameco is unaware of any data which contradicts or undermines this long-standing conclusion which was based upon a substantial body of scientific literature. The uranyl peroxide yellowcake produced at Crow Butte is dried with a dryer oil temperature of approximately 230°C. During inspections at Crow Butte, NRC has repeatedly evaluated both the historic results of the exposure program and the appropriateness of the selected DAC, and has confirmed the adequacy of the bioassay program. As such, the current solubility classification remains valid and a revised lower limit of detection for measuring uranium in air is not required.

As a matter of information, Cameco has attached a copy of recently published paper with solubility data from Crow Butte and Smith Ranch yellowcake entitled "Solubility of Radionuclides in Simulated Lung Fluid", presented on August 18, 2010 at the Uranium 2010 Conference in Saskatoon, Saskatchewan, Canada. Please note that the results presented in the Uranium 2010 paper are based upon one randomly selected Crow Butte sample and one randomly selected Smith Ranch sample. That study assigned Type F to both Crow Butte and Smith Ranch yellowcake. For additional detail on the two-term exponential model used to define dissolution times please refer to the attached paper.

Figures 1 and 2 present the dissolution curves (% undissolved as a function of time) for the Crow Butte and Smith Ranch samples respectively, aligned with the ICRP 71 F/M/S classification system (see ICRP 71 Appendix D).

Figure 1: Dissolution of Crow Butte Samples

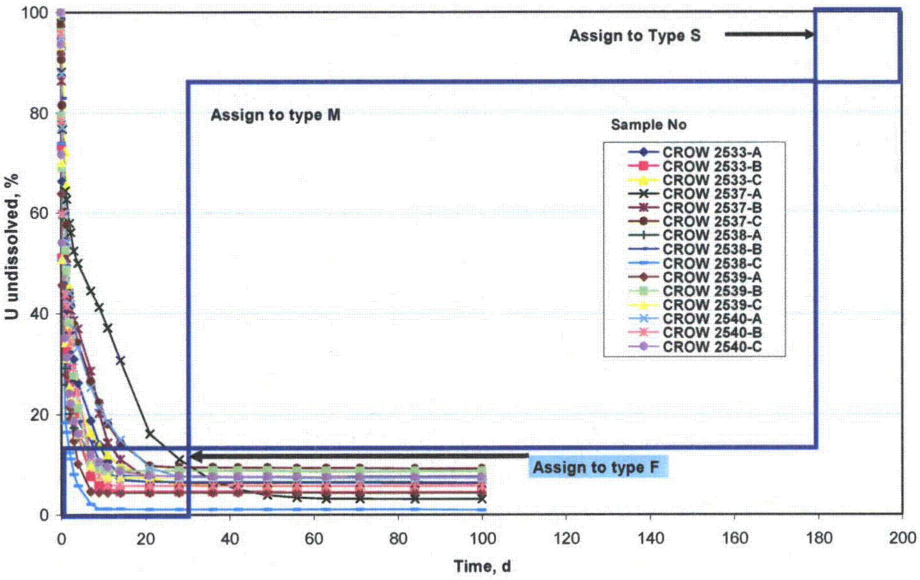
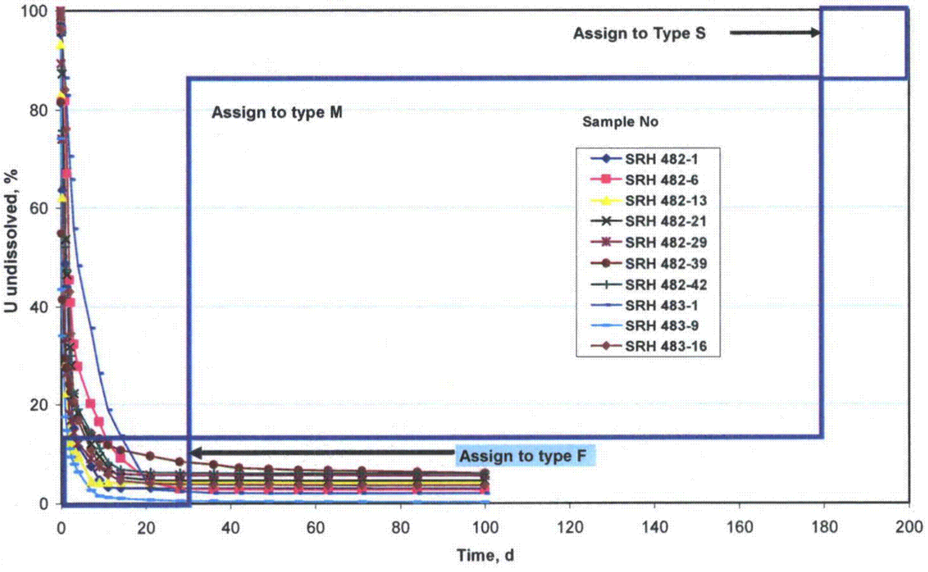


Figure 2: Dissolution of Smith Ranch Samples



Using the ICRP 30 model, the experimental results describe above were also interpreted in the context of the days, weeks, and years (D/W/) solubility classification. In terms of the D/W/Y classification, the majority of the samples were classified as Type D with a very small component of Class W material:

Site	# of samples	F1 Avg. (%)	T1 Avg. (days)	F2 Avg. (%)	T2 Avg. (days)	ICRP 30 % D	ICRP 30 % W	ICRP 30 % Y	ICRP 71
Crow Butte	15	68.6	0.6	31.4	34.5	89.5	9.5	0.5*	All F
Smith Ranch	10	86.7	1.1	13.3	47.8	95.7	4	0.3*	All F

**The analyst expressed concern that the two samples with a Class Y component (one from each plant) represent an extremely small amount of undissolved material remaining after 100 days and for that reason may reflect statistically unreliable data.*

It is clear that yellowcake produced at Crow Butte is quite soluble and should be assigned as 100% Class D for purposes of the 10 CFR 20, Appendix B, Table 1 DAC for natural uranium. Despite the <10% Class W component, the yellowcake is considered soluble and meets the definition of "low fired yellowcake". The definition of "low fired yellowcake" used in Regulatory Guide 8.22 is defined in NUREG 0874 as:

Low Temperature Drying	Fractional Composition	Dissolution Half Time (days)	Inhalation (Solubility) Class
Short Lived Component	0.61	0.8	D
Medium Lived Component	0.39	39	W

From this, 39% of the uranium product may have a dissolution half time up to 39 days (i.e. a significant Class W component) and still be considered 100% Class D soluble "low fired yellowcake" for application of Reg. Guide 8.22.

Finally, the default Type D ALI and DAC values for U natural found at 10 CFR 20 Appendix B, Table 1, are roughly 3.5 times more conservative than the results calculated by Cameco using the solubility parameters determined by the studies and the Human Health Respiratory Tract Model in ICRP 66. This means that use of the default DAC value will overestimate dose by a factor of 3.5 and that adjustment of the DAC value pursuant to 10 CFR 20.1204(c) may be warranted.

C. Section 5.8.3.1, Airborne Uranium Particulate Monitoring

2. In CBR's May 12, 2009 letter (ML091470119) responding to NRC staff's Request for Additional Information (ML083660060), it did not provide sufficient information to justify the use of gross alpha counting for determining occupational internal dose from sources other than radon and its daughters. This method does not account for potential radiation from other alpha emitters, as well as the potential for the presence of beta-gamma emitting daughters. Please provide technical justification for relying on gross alpha counting alone for determining occupational internal dose from sources other than radon and its daughters.

***Response**: In accordance with 10 CFR §20.1204(g), nuclides may be ignored in a mixture in air if the total activity in the mixture is used to determine compliance with §20.1201 and §20.1502(b), any nuclides ignored are <10% of the mixture, and the sum of all nuclides ignored are <30% of the mixture.*

Other Alpha Emitters

Studies performed in the late 1970s and early 1980s of radionuclide mobilization from several ISRs and subsequent measurements at operating ISRs indicate a relatively small portion of the uranium daughter products in the ore body are actually mobilized by the lixiviant. (See e.g., Brown, S. 1982, Radiological Aspects of Uranium Solution Mining, In: Uranium, 1, 1982, p. 37-52, Elsevier and Brown, S, 2007, Radiological Aspects of In Situ Uranium Recovery. American Society of Mechanical Engineers, Proceedings of 11th International Conference on Environmental Management, Bruges, Belgium, *September; ASME Press, New York, NY, ISBN 0-7918-3818-8). In these studies, Th-230 appeared to equilibrate, with very little removed from the host formation by the process. The majority of the mobilized radium-226 (80-90 percent), estimated to be approximately 5 to 15 percent of the calculated equilibrium radium in the host formation, followed the calcium chemistry in the process and resulted in radium carbonates/sulfates in the calcite byproduct waste streams. Little, if any, lead-210 was mobilized, as the lead carbonate complexes formed in situ are virtually insoluble in the lixiviant processes studied.*

In addition, the ion exchange (IX) resin used in ISR facilities is specific for removal of uranium. The very small amount of thorium-230 that is mobilized in the lixiviant is not removed by the IX resin and returns to the formation with the lixiviant. For that reason, it is not present in the process downstream of the IX columns (e.g., elution, precipitation, and drying circuits). Accordingly, the "nuclide mix" that can potentially become airborne in the precipitation, drying and packaging areas of a modern ISR is expected to be almost exclusively Unat.

Cameco collected air sample data at Smith Ranch during February and July 2010 to ascertain if the progeny relative to the uranium parent in air is sufficiently low to allow application of 10 CFR §20.1204(g). Due to the clean nature of Cameco operations at Smith Ranch, very little radioactivity was present on any of the filters. For uranium, the most activity on any filter was reported as 23 pCi (Dryer Round 2), with an associated airborne concentration of 2.4E-12 uCi/l (< 1% of the Class D natural uranium DAC). For almost all progeny measurements, results were in fact at or below the reported minimum limit of detection (MDC) by the laboratory. Although numerical values were reported for the progeny, most are reported as negative #s (-) implying the actual sample result was less than the MDC. That is, “non-detectable”.

For that reason, Cameco performed isotopic analysis of fresh yellowcake samples from Smith Ranch and Crow Butte to demonstrate applicability of 10 CFR §20.1204(g). If the nuclides are not present in the fresh yellowcake they will not be present in air samples taken at the facilities. The fresh yellowcake results include:

Site (Laboratory)	***Sample Receipt***	***Total Uranium***	***Thorium-230***	***Radium-226***
Smith Ranch (Energy Labs)	*5/26/10*	*493,000 pCi U/g sample*	*26 pCi / gram sample*	*20.8 pCi / gram sample*
Crow Butte (Energy Labs)	*8/9/10*	*540,000 pCi U/ g sample**	*30 pCi / gram sample (reported as 1.1 Bq/ gram)*	*86 pCi / gram sample (reported as 3.2 Bq / gram*

**Assuming 79.54% U composition*

As an example of the calculation, the activity of the Smith Ranch yellowcake was:

- *7.36 E5 mg Unat / kg sample X E-3 kg sample/g sample = 7.36 E2 mg Unat / g sample*
- *7.36 E2 mg Unat / g sample X E-3 g Unat/ mg Unat = 7.36 E-1 g Unat / g sample**
- *7.36 E-1 g Unat / g sample X 0.67* uCi Unat / g Unat = 4.93 E-1 uCi Unat / g sample*
- *4.93 E-1 uCi Unat / g sample X E6 pCi / uCi = 4.93 E5 pCi Unat / g sample = 493,000 pCi Unat / gram of sample*

** Specific activity of natural uranium per 10 CFR 20, Appendix B*

Relative to the radium-226 and thorium-230 results, the radionuclide composition of the samples are >99.9% uranium, justifying application of 10 CFR §20.1204(g).

<u>Presence of Beta and Gamma-emitting Daughters</u>

It must be recognized that in-growth of the short-lived beta emitters Th-234 and Pa-234 from the freshly extracted uranium product takes approximately 4 months to reach equilibrium. Accordingly, *very little, if any Th-234 or Pa-234 would be expected to be present in the active processing areas at the Crow Butte Facility. Additionally, the DACs for these two nuclides are several orders of magnitude higher than for U natural. Controlling to the DAC for U natural ensures any contribution in the mixture from these two beta emitters will meet the 10% exclusion allowance in 10 CFR §20.1204(g). By way of validation, see response D.1., immediately below.*

D. Section 5.8.6.1, Surveys for Surface Contamination

1. Please address how the potential for the presence of beta-gamma emitters are addressed in CBR's contamination control program.

***Response**: In addition to gamma surveys, Regulatory Guide 8.30 recommends that beta surveys of operations that involve direct handling of large quantities of aged yellowcake be performed to ensure that extremity and skin exposures are within limits. This includes plant areas associated with precipitation, dewatering (belt filter) and drying/packaging. Beta contamination surveys are performed in these plant areas initially and whenever procedural and/or equipment changes may increase risk of beta contamination. These surveys are typically performed with a Ludlum Model 3 survey meters and model 44-38 GM detectors. This probe is a thin walled halogen quenched GM tube with movable beta shield with manufacture reported backgrounds of 20 cpm window closed and 25 cpm open. These surveys will be performed near the surface of the material (e.g., within 10 cm) to represent beta exposure rates to workers' hands and skin during the handling of the material. Any beta exposure rate evaluations for these operations that are performed in lieu of instrument surveys will use the information provided in Regulatory Guide 8.30, Figures 1 and 2.*

However, it should be recognized that there are no mechanisms by which the beta emitters Thorium-234 or Proactinium-234 can be separated from their alpha emitting uranium parents. For that reason there cannot be contamination in the absence of detectable alpha. The maximum possible beta activity exists when the beta emitting daughters are at equilibrium with the uranium, approximately 4 months after mining. Cameco typically drums yellowcake as it is produced (providing shielding from any beta emitter in growth) and transports all yellowcake offsite as soon as a full shipment is accumulated such that there should not be any "aged yellowcake" on site. For that reason, the Cameco contamination control program relies primarily upon alpha surveys and there is no circumstance in which beta exposure can occur.

This process perspective has been verified by extensive beta/gamma surveys that have been performed throughout the Crow Butte and Smith Ranch plants including in yellowcake areas. Beta/gamma surveys were conducted at Smith Ranch on May 26, 2010 and at Crow Butte on June 17, 2010. Over 35 locations were surveyed at each plant including in the immediate vicinities of IX and elution columns, precipitation tanks, filter press and dryer areas. At each location, a pair of measurements were made, one with the GM probe beta shield open (beta + gamma) and closed (gamma only). With the exception of the belt filter at Crow Butte, at no location in either plant was the beta and gamma result in mR/hr greater than the gamma only result. This demonstrates no measurable contribution to exposure from beta emitters, and validates the assumption that there are no appreciable beta emitting uranium progeny in fresh yellowcake or in intermediate ISR products. The belt filter is one of the areas addressed above in the first paragraph of this response.

D. Section 5.8.6.1, Surveys for Surface Contamination

2. Please provide the frequency of contamination surveys in the operating areas of the facility.

***Response**: The frequencies are presented in Table 5.8-16.*

D. Section 5.8.6.1, Surveys for Surface Contamination

3. In Section 5.8.6.1 of the TR, CBR states that an action level of 25 percent of the limits from RG 8.31 is used for clean areas. Please provide details of survey equipment used for this purpose and demonstrate the lower limit of detection of these instruments.

***Response**: The reader is directed to the survey equipment described in Section 5.8.6.3. The Ludlum 43-5 and 43-65 (or equivalent) alpha scintillation probes have alpha efficiencies of 13% and 17% respectively (rated for plutonium-239) and a background of ≤3 cpm. The Ludlum 44-38 beta-gamma GM probe (or equivalent) has a sensitivity of 1200 cpm/mR/hour (rated for Cs-137) and will detect any significant beta or gamma contamination.*

E. Section 5.8.6.2, Surveys for Contamination of Skin and Personal Clothing

Please address how the potential for the presence of beta-gamma emitters are addressed in CBR's personnel contamination control program.

***Response**: Please see the response to D.1., above. The Crow Butte contamination control program relies primarily upon alpha surveys.*

F. Section 5.8.6.3, Surveys of Equipment Prior to Release to an Unrestricted Area

1. Please address how the potential for the presence of beta-gamma emitters are addressed in CBR's program for releasing items for unrestricted use.

***Response**: All equipment and materials released from the Restricted Area are surveyed for alpha contamination and for beta gamma contamination. The release limits for beta gamma contamination are 0.2 mrad average and 1.0 mrad maximum at 1 cm as required by "Guidelines for Decontamination of Facilities and Equipment Prior to Release for Unrestricted Use or Termination of Licenses for Byproduct, Source, or Special Nuclear Material" dated May 1987 (License Condition 9.8). A Ludlum Model 3 survey meter with a Ludlum 44-38 probe (or equivalent) will be used. Section 5.8.6.3 has been revised to reflect the release limits.*

F. Section 5.8.6.3, Surveys of Equipment Prior to Release to an Unrestricted Area

2. In CBR's May 12, 2009 letter (ML091470119) responding to NRC staff's Request for Additional Information (ML083660060), it did not provide sufficient regulatory justification to relieve it from complying with License Condition (LC) 9.12. Specifically, LC 9.12 requires compliance with Regulatory Guide (RG) 8.31. CBR has not proposed a program that is consistent with RG 8.31 regarding the qualifications of personnel releasing items for unrestricted use. Please provide qualifications of personnel releasing items for unrestricted use that are consistent with LC 9.12 and RG 8.31.

***Response**: The text has been clarified as to the qualifications of personnel releasing items for unrestricted use.*

G. Section 5.8.7, Airborne Effluent and Environmental Monitoring Program

1. Please specify radiological monitoring that CBR will perform during operations to confirm its licensing basis and the validity of calculations used for estimating public and occupational exposures for all aspects of its operations.

   Although calculations are allowed in meeting the requirements of 10 CFR 1301/1302, the NRC staff expects that CBR will perform monitoring to such an extent as to be able to confirm its licensing basis and the validity of calculations used for estimating effluent concentrations and calculating dose for compliance with 20.1301/1302. This concept applies to point sources as well as diffuse sources such as radon released in the wellfield. Compliance with 10 CFR 20.1501 in regards to occupational dose should be approached in the same manner.

Monitoring proposed by CBR does not appear to take into account all sources of potential exposure to workers and members of the public. Examples include effluent discharged from the facility, header houses, and wellfields.

More specifically, it is not clear that occupational exposures due to radon and its progeny are considered for all aspects of its operations throughout the permit area.

Please provide a more detailed description of CBR's radiological monitoring program that can be used to confirm its licensing basis and the validity of calculations used for estimating public and occupational exposures for all aspects of its operations.

***Response**: The radiological monitoring CBR performs to confirm its licensing basis and the validity of calculations is detailed in Section 5 of the license renewal application. The license renewal application also presents historic results. In addition, monitoring results are provided to the NRC in the Semiannual Effluent Monitoring Report which includes a table entitled "Dose to Public Calculations". During inspections, the NRC uses the semiannual report to review both the licensing basis and the validity of the underlying calculations. Neither the monitoring nor the validity of calculations has been called into question during the NRC inspections.*

CBR uses the results of process area and environmental monitoring for direct radiation (see Sections 5.8.2.1, 5.8.7.2 and 5.8.7.7), air particulates (see Sections 5.8.3.1, 5.8.3.2 and 5.8.7.3) and radon (see Section 5.8.7.4) to estimate occupational exposure and dose to the public from licensed operations. A summary is presented in Table 5.8-5. The monitoring results are used to calculate the in-plant TEDE (Section 5.8.3.3), the natural uranium and pre-natal exposure (Sections 5.8.4.1 and 5.8.4.2 respectively), and the TEDE for nearby residents (Table 7.12-3).

<u>Dose to Public</u>

As described in Section 7.12.5, the MILDOS Area simulation gave a maximum TEDE of 31.7 mrem/year for a resident located at Receptor #15. Receptor #15 (see Figure 7.12-7) is located at the permit boundary approximately 0.25 miles northeast of the plant site, at the same location as Air Monitoring Station AM-8. The 2009 "Dose to Public Calculation" (noted above), based upon data collected at AM-8, showed an actual TEDE of 17.00 mrem/year to a resident, or approximately 54% of the MILDOS simulation. Similarly, the MILDOS Area simulation gave a TEDE of 19.9 mrem/year for a resident located at Receptor #20. Receptor #20 (see Figure 7.12-7) is located within a wellfield approximately 1.3 miles north-northeast of the plant site, at the same location as Air Monitoring Station AM-2. The 2009 "Dose to Public Calculation" (noted above), based upon AM-2 data, showed an actual TEDE of 2.12 mrem/year to a resident, or approximately 11% of the MILDOS simulation.

As such, CBR monitoring demonstrates the conservatism of the MILDOS Area simulation at the location of highest dose to an identified public receptor. Similar to the MILDOS Area simulation, the monitoring and resulting Dose to Public TEDE includes the dose from uranium, radium-226, lead-210, radon-222 and gamma from the diverse, albeit limited sources in and around the plant, and provides consideration of radon and its progeny.

<u>Occupational Dose</u>

The current monitoring conservatively brackets the occupational dose to workers between the plant and the site boundary. The MILDOS Area simulation described in Section 7.12.5 may be used to overestimate worker dose in this area. The MILDOS Area simulation considers radon and its progeny. Examination of Figure 7.12-7 reveals a cluster of receptors (3, 11 and 21) located in wellfield but with minimal influence from plant emissions. Taking the worst-case of these TEDEs (Receptor 11), and multiplying by 2000 hours/8760 hours, gives 1.7 mrem/year worker exposure from wellfield sources. It should be noted that this value includes exposure from vegetation and meat consumption that would not occur for a worker.

Receptors 14, 15, 18 and 20 may also be used to simulate occupational dose to workers where both wellfield and plant contributions are present. Taking the worst-case of these TEDEs (Receptor 15), and multiplying by 2000 hours/8760 hours, gives 7.2 mrem/year worker exposure from wellfield and plant sources. As noted above, this value includes exposure from vegetation and meat consumption that would not occur for a worker. The monitoring performed at AM-8 and AM-2 again demonstrates the conservatism of the MILDOS Area simulations.

G. Section 5.8.7, Airborne Effluent and Environmental Monitoring Program

2. In CBR's May 12, 2009 letter (ML091470119) responding to NRC staff's Request for Additional Information ML083660060), it did not provide sufficient information to demonstrate compliance with 10 CFR 20.1301(b) and 10 CFR 20.1302(b). CBR appeared to use arbitrary downwind receptor points for its calculations. In terms of public dose, it is not clear that CBR determined who or what group receives the highest exposure consistent with NUREG-1736. Please provide an analysis of public dose consistent with open issue G(1) above and NUREG-1736.

***Response**: CBR compliance with 10 CFR 20.1301(b) and 10 CFR 20.1302(b) have been routinely evaluated by NRC through review and issuance of the current license and review of the Semiannual Radiological Effluent and Environmental Monitoring Reports. Similarly, the downwind receptor points were reviewed and approved by the NRC and determined appropriate.*

Section 2.5.5 states: "the predominant air pollutant dispersion would be towards the north to north east. The next most common directions would be towards southwest to south-southwest". Figure 2.9-1 shows that two monitoring stations are located to the north-northeast and another to the southwest to south-southwest. As such, the receptor locations used in the calculations are not arbitrary.

As noted in response G.1., a resident located approximately 0.25 miles northeast of the plant site receives the highest exposure.

H. Section 5.8.7.2, Radon – Please provide information on how radon daughters are factored into determining compliance with 10 CFR 20.1301/1302 during operations.

***Response**: CBR uses the results of environmental monitoring for direct radiation (see Section 5.8.7.2 and 5.8.7.7), air particulates (see Section 5.8.7.3) and radon (see Section 5.8.7.4) to estimate dose to the public from licensed operations. These assessments are provided in the Semiannual Effluent Monitoring reports which are routinely reviewed as part of NRC inspections. Issues have not been identified. For additional detail see the response to G. 1., above.*

II. Confirmatory Items

A. Section 6.3, Removal of Process Buildings and Equipment – Please confirm that the beta-gamma limits of the Guidelines will be applied to the release of items for unrestricted use.

***Response**: In Section 6.3 CBR references the guidelines in their entirety, without qualification, and notes the NRC authority to review and approve the survey.*

III. Administrative items

A. General – change the verb "would" to "will" in the TR when describing the applicant's program unless the action will be contingent upon some other action.

***Response**: Upon issuance of the final License Renewal application, where appropriate, "would" will be changed to "will".*

B. Section 2.5, Meteorology - Table 2.5-15, "Joint Frequency Distribution", indicates that the data is comprised of a summary of meteorological conditions over a one-year period but does not indicate the actual time period. Please indicate the one-year time period that this data represents.

***Response**: The actual time period will be indicated in the footnotes to Table 2.5-15.*

C. Section 4.2.1.2, Secondary Water Sources – In Section 4.2.1.2 of the TR, the applicant discusses how liquid waste from the laboratory will be disposed. Please provide information on amounts of laboratory waste that contains hazardous material and how this waste is dispositioned.

***Response**: Liquid waste from sink drains in the laboratory is piped directly into the liquid waste disposal system and disposed in the evaporation pond and deep disposal well. The estimated flow is 3,000 gallons per month. The nonhazardous liquid wastes include sample discards, lab solutions, lab cleanup and dishwashing.*

SOLUBILITY OF RADIONUCLIDES IN SIMULATED LUNG FLUID

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ABSTRACT

The objective of this work was to assign FMS (fast, moderate and slow) absorption types to site-specific radionuclides found at various Cameco facilities. Kinetics studies of dissolution of various uranium-bearing samples in simulated lung fluid were carried out. Dissolution parameters were determined and the samples were assigned to the FMS absorption types. A software program was developed for the determination of dissolution parameters. The assignments were based on criteria established in the International Commission on Radiological Protection (ICRP) Publication 71. The assignments were used to determine the internal radiation dose for workers.

MET SOC
CIM ICM
Uranium 2010 - "The future is U"
Proceedings of the 3rd International Conference on Uranium
40th Annual Hydrometallurgy Meeting
Saskatoon, Saskatchewan, Canada
Edited by E.K. Lam, J.W. Rowson, E. Özberk

INTRODUCTION

One of the regulations of the Canadian Nuclear Safety Commission (CNSC) requires that an internal dose component be assigned for personnel working at Cameco sites in Canada. This approach was expanded to Cameco's US operations. Simulated lung fluid (SLF) solubility experiments are carried out for the various types of uranium-bearing materials present at the sites in order to produce site-specific solubility data. Without the site-specific solubility data, more conservative default values for the various uranium-bearing materials would have to be used.

A critical review of techniques for studies of solubility of uranium compounds in simulated lung fluid was described in a previous report [1]. The method used in this study was selected after a thorough analysis of the advantages and disadvantages of the existing techniques described in the literature.

For the purpose of calculating the radiation doses from radionuclides deposited in the lung by inhalation, uranium compounds were first divided into three solubility classes based on retention times in the lungs, according to ICRP Publication 30 (1979) [2]:

- Class D compounds, less than 10 days
- Class W, 10-100 days
- Class Y, greater than 100 days.

Further conditions were added, and, in the ICRP Publication 71 (1995) [3], the clearance of contaminating material from the lungs was treated as a result of two processes, namely, particle transport to the gastrointestinal tract and absorption into body fluids. For simplicity, lung retention (the amount of material that was not dissolved) for *in vitro* dissolution experiments was considered in the absence of particle transport simulating respiratory tract absorption. Subsequently, three new absorption types F, M or S (fast, moderate and slow) were created. Assignments to types F, M and S were carried out using the following criteria. If the retention was below 13% in 30 days, the material was assigned to type F. If the retention was in the range 13-87% between 30 and 180 days the material was assigned to type M. If the retention was above 87% in 180 days, or later, the material was assigned to type S. These categories correspond approximately to classes D, W and Y described in ICRP Publication 30 (1979).

During 2000-2005, an in-house method was developed to measure solubility parameters for uranium concentrate samples and the uranium site-specific, process-stream samples and aerosol samples collected at the Port Hope conversion facility (PHCF) and Blind River refinery (BRR). An additional series of SLF solubility experiments were carried out for uranium and other radionuclides-bearing materials present at the Key Lake (KLO), McArthur River (MCA) and Rabbit Lake (RLO) uranium facilities in order to produce site-specific solubility data. The data obtained were used to assign to the solubility types according to the International Committee on Radiological Protection (ICRP) Publication 71 requirements and to assign an internal dose component for personnel working at these stations. In 2006, experiments were carried out to study the solubility of 30 additional samples collected at the PHCF, Zircatec Precision Industries (ZPI; since renamed Cameco Fuel Manufacturing), Cameco technology development (CTD; since renamed Innovation & Technology Development – Research Centre) and Blind River refinery (BRR).

In 2009, a request was received from Cameco's safety, health, environment and quality (SHEQ) department to carry out similar lung fluid dissolution studies for uranium concentrate samples collected at the Crow Butte (CBO), Smith Ranch-Highland (SRH) and Rabbit Lake (RLO) operations.

The objective of this work was to assign the site-specific uranium compounds to absorption types according to the International Committee on Radiological Protection (ICRP) Publication 71 criteria.

For the sake of brevity, only a small part of the SLF studies of radionuclides are presented in this publication.

EXPERIMENTAL

The experimental work included:

1) obtaining samples from Cameco sites
2) sieving and separation of samples to obtain the inhalable particle size <20 µm
3) analysis of samples for total uranium (carried out at BRR analytical services)
4) studies of the dissolution kinetics in SLF
5) calculation of kinetics parameters and assignment of samples to FMS absorption types

The Samples Used in the Experiments

Cameco Port Hope conversion facility (PHCF) process stream samples and Cameco concentrate samples obtained from mine sites were used in experiments.

The PHCF process stream samples included:

(i) Uranium tetrafluoride
(ii) Uranium dioxide, UO_2 (ceramic)
(iii) Uranium trioxide, UO_3
(iv) Ammonium diuranate, ADU

The results of total uranium analyses in PHCF process stream samples are presented in Table 1.

Table 1 - Cameco Port Hope conversion facility process stream samples used in the experiments

Compound	Formula	Sample No. Comments	Total Uranium in 50 mg Sample (mg)
Uranium tetrafluoride calcined	UF_4	Calciner C #000311	38
Uranium dioxide ceramic	UO_2	Lot#200DL154	44
Uranium trioxide	UO_3	Autosampler Lot#275492	42
Ammonium diuranate	$(NH_4)_2U_2O_7{\bullet}XH_2O$		38

Description of the Cameco concentrate samples, used in the experiments, including the results of total uranium analyses, are presented in Table 2.

Table 2 - Cameco concentrate samples used in the experiments

Concentrate Sample / Location/ Experiment year	Total Uranium in 50 mg Sample (mg)
Crow 2009, Crow Butte Operation, 2009	38.4
KLO 2005, Key Lake Operation, Packaging Area, 2005	39.6
RLO 2009, Rabbit Lake Operation, 2009	34.0
SRH 2009, Smith Ranch Operation, 2009	39.5

Reagents

The reagents used are presented in Table 3.

Table 3 - The reagents used in the solubility experiments

Chemical/Description	Supplier
Nitric acid (double-distilled), HNO_3	Seastar Chemicals Inc.
Hydrochloric acid, concentrated (35-37% assay), HCl	Fisher Scientific
Magnesium chloride hexahydrate, $MgCl_2•6H_2O$	Fisher Scientific
Sodium chloride, NaCl	Fisher Scientific
Potassium chloride, KCl	Fisher Scientific
Sodium hydrogen phosphate heptahydrate, $Na_2HPO_4•7H_2O$	Fisher Scientific
Sodium sulphate, Na_2SO_4	Fisher Scientific
Calcium chloride dihydrate, $CaCl_2•2H_2O$	Fisher Scientific
Sodium acetate trihydrate, $NaH_3C_2O_2•3H_2O$	Fisher Scientific
Sodium hydrogen carbonate, $NaHCO_3$	Fisher Scientific
Sodium citrate dihydrate, $Na_3H_3C_6O_7•2H_2O$	Fisher Scientific
Deionized water	Laboratory Supply

Simulated Lung Fluid

The composition of SLF solutions used in the experiments is illustrated in Table 4. This formulation was suggested by Moss in 1976 [4]. The selection of the lung fluid was based on the analysis of the information available in the literature [5].

The electrolyte compositions of actual human lung fluid and the selected simulant were almost identical. Moss suggested replacing the protein components of actual lung fluid by an ionically equivalent amount of citrate in the simulant. Actual lung fluid proteins are poorly characterized and generally not available in large quantities, and substitute proteins hinder filtration and promote bacterial growth in solutions. Phospholipids, also known to be present in trace amounts in actual lung fluid, were not included in the simulant for the same reasons. The previous studies did not indicate any effect of organic ingredients on the dissolution rate of uranium yellow cake samples.

Simulated lung fluid was prepared in ten-litre batches by addition of ten-times the amounts of the components listed in Table 3 into 1 L of distilled water in a 10-L polyethylene container. Each reagent dissolved by continuous stirring and only then the next was added. The aqueous solution obtained was diluted with distilled water up to the 10-L mark. The solution was filtered and saturated with air/5% CO_2. Finally, the pH of the solution was adjusted to 7.3-7.4 by addition of small volumes of 1N hydrochloric acid, HCl. The pH of the solution was checked every day and maintained at 7.4 by addition of HCl as needed.

Table 4 - Composition of the additives in 1 L of aqueous SLF solution (O.R. Moss, 1976)

Name	Formula	Mass (g)
Magnesium chloride hexahydrate	$MgCl_2•6H_2O$	0.2033
Sodium chloride	NaCl	6.0193
Potassium chloride	KCl	0.2982
Sodium hydrogen phosphate heptahydrate	$Na_2HPO_4•7H_2O$	0.2680
Sodium sulphate	Na_2SO_4	0.0710
Calcium chloride dehydrate	$CaCl_2•2H_2O$	0.3676
Sodium acetate trihydrate	$NaH_3C_2O_2•3H_2O$	0.9526
Sodium hydrogen carbonate	$NaHCO_3$	2.6043
Sodium citrate dihydrate	$Na_3H_3C_6O_7•2H_2O$	0.0970

Instrumental

The extracted uranium concentration in simulated lung fluid was determined using inductively coupled plasma-mass spectrometry (ICP-MS).

An ATM sonic sifter was employed to obtain samples with particle sizes less than 20 µm.

Experimental Circuit

Experiments were carried out in a circuit previously employed for the studies of the kinetics of uranium dissolution in SLF (Figure 1). The experimental circuit was housed in a wooden box to prevent any contamination by spurious uranium contaminant that might have been present in the laboratory environment. The box was insulated and the temperature was maintained at 37°C using a thermostatically controlled heater-fan. Access to the different parts of the apparatus was through wooden and Plexiglas double doors. The SLF reservoir and the sample collection bottles were sealed with plastic caps. All other components in contact with the solution were plastic. Other materials, such as metal and glass, were avoided because of potential corrosion, contamination and adsorption problems.

The 50-mg concentrate samples (particle size <20 µm) were placed between two glass fiber filters in 47-mm polypropylene filter holders used as extraction cells. The outlet sides of the extraction cells were connected by Teflon tubing to 500-mL polypropylene sample collection bottles.

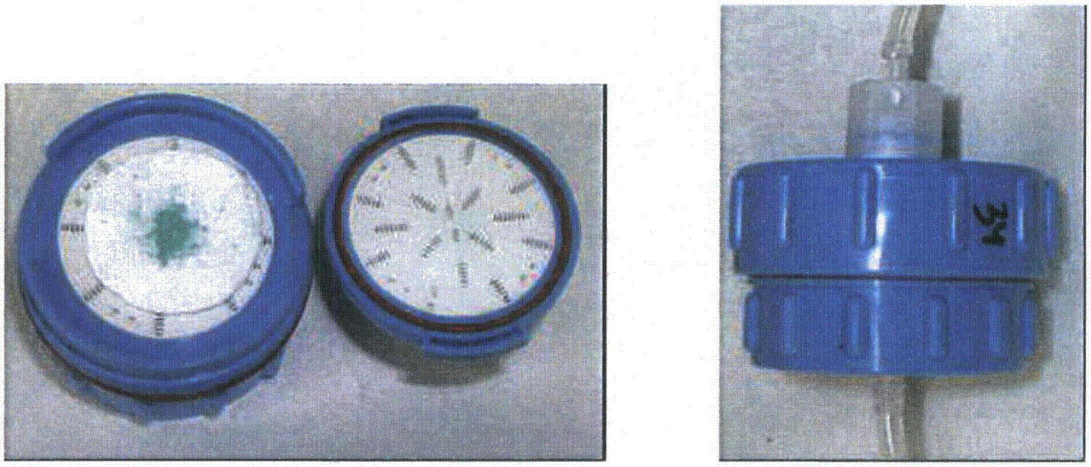

Figure 1 - A filter holder used as an extraction cell in experiments

The required quantities of the original samples were estimated based on the concentrations of uranium in the sample and the volumes of SLF used for the dissolution. The required volumes of SLF were estimated based on the flow rates, applied over 100 days of extraction with consideration of the detection limits for the uranium and with the assumption that the uranium contained in the samples had a low solubility in SLF. The SLF was passed through the sample at predetermined flow rates.

The SLF was supplied to the extraction cells from two 8-L vessels using a forty-channel peristaltic pump. A third 10-L vessel was used to continuously supply fresh SLF using a two-channel peristaltic pump. The solution was saturated with a 5% mixture of CO_2 in air to simulate (to a degree) the oxidizing conditions in the lung.

The starting flow rates were significantly higher to prevent oversaturation (and possible re-precipitation) of uranium at the beginning of the experiments, when the concentrations of the uranium was high. The flow rates were gradually decreased from 1 mL/min to 0.05 mL/min with the decrease of uranium concentrations in the sample in order to obtain the concentrations of dissolved elements in SLF, which would be sufficient for measurements. The volumes of simulated lung fluid samples with dissolved uranium gradually increased over the course of extraction.

The samples of SLF, after passing through the extraction cells, were collected, acidified with double-distilled nitric acid (to prevent precipitation of elements) and submitted to the PHCF analytical services for determination of the uranium concentration by ICP-MS.

An uninterrupted power supply line was installed to the circuit to ensure continuous operation.

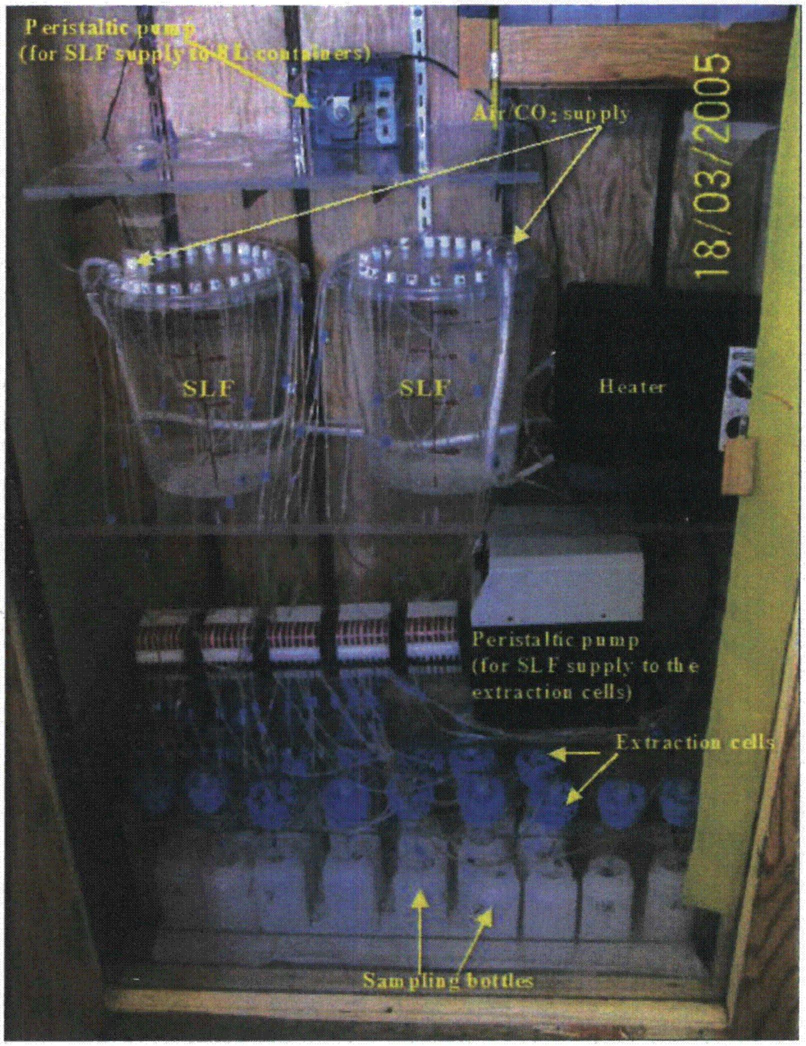

Figure 2 - The circuit for the studies of solubility of radionuclides in SLF

RESULTS AND DISCUSSIONS

Dissolution Kinetics and Dissolution Parameters

Dissolution half-times for a composite aerosol sample were calculated using the two-exponential model and the following Equation (1), described previously, where percentage of undissolved uranium is expressed as [6]:

$$M/M_0=f_1\exp(-0.693t/T_1) + f_2 \exp(- 0.693t/T_2) \quad (1)$$

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

$f_1 + f_2 = 100\%$

A software program, developed at the research centre, was employed for the determination of dissolution parameters.

The program uses a Simplex algorithm to minimize the functional F calculated as the sum of squared differences between the experimental and theoretical values characterizing the uranium content (fraction of dissolved or undissolved uranium). To ensure that the global minimum is found, the minimization calculations are repeated several hundred times using Monte Carlo technique to vary the initial conditions. It is assumed that the global minimum is the set of optimized parameters corresponding to the smallest functional found in the series of these 200 to 500 minimizations. The procedure was tested several times and it was confirmed that 200 repetitions are sufficient to locate the global minimum.

After the global minimum is found, the Hessian matrix consisting of the second derivatives of F with respect to all the free parameters involved in the model is calculated. The calculations are performed using analytical expressions for the second derivatives (as opposed to numerical differentiation in most of the programs such as MathCAD or Mathematica). It was checked for each derivative that the analytical expressions were consistent with the numerically differentiated. The standard deviations of parameters were calculated from the inverse Hessian matrix using the MSE (Mean Square Error) 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).

Absolute values of standard errors for parameters are given in parenthesis. 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 equation (7) allows one to determine the rapid (f_r) and slow (f_s) fractions and their corresponding half-times, T_r and T_s. The equation $S = \ln2/T$, allows the half-time T to be transformed into dissolution rate constant S (or S_r and S_s, respectively). Parameters f_r, S_r and f_s, S_s can be used in a LUDEP program to calculate the dose intake according to ICRP Publication 66.

A program was developed at Cameco research centre for calculation of dissolution parameters using non-linear regression analysis.

Dissolution kinetics for hypothetic two-component sample containing fraction f_1 with dissolution half-time T_1 and fraction f_2 with dissolution half-time T_2 is illustrated in Figure 3.

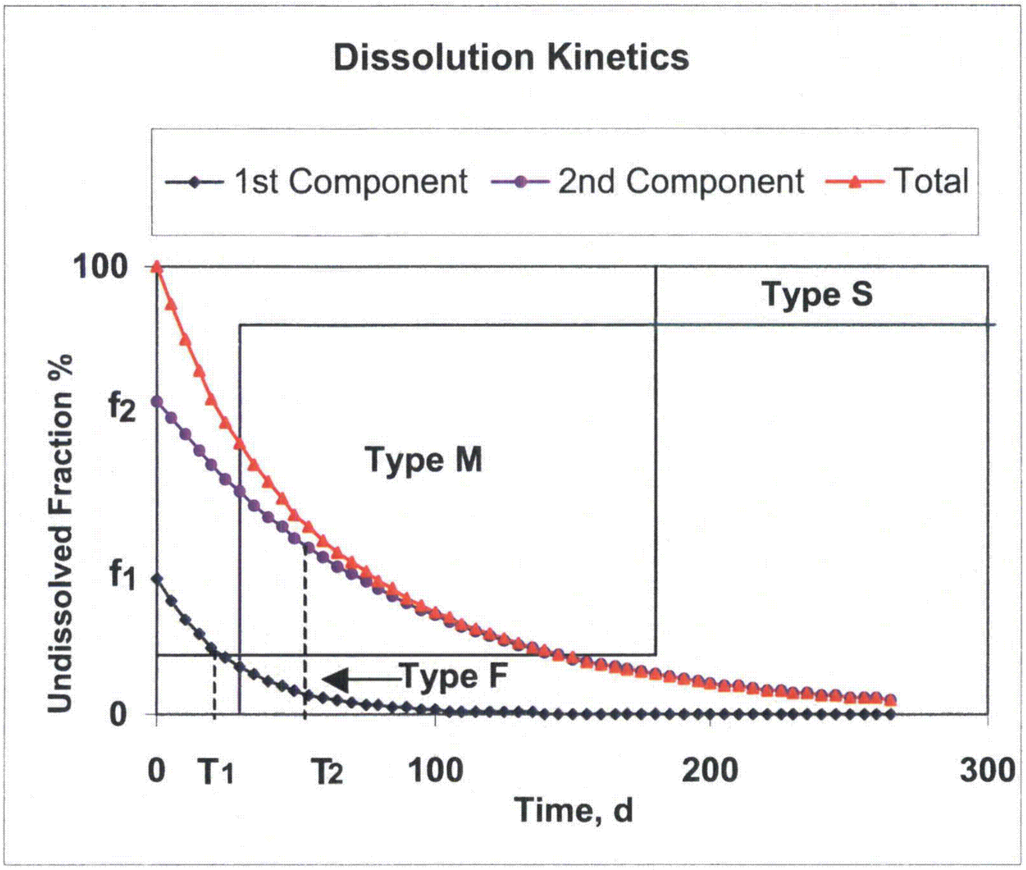

Figure 3 - Dissolution kinetics for two-component sample containing fraction f_1 with dissolution half-time T_1 and fraction f_2 with dissolution half-time T_2

In the ICRP 71 respiratory tract model (1995) [7], the clearance of contaminating material from the lungs is treated as a result from the sum of two processes, namely, the particle transport to the gastro-intestinal tract and the absorption into body fluids. For simplicity, for *in vitro* dissolution experiments, lung retention (the amount of material that has not dissolved) is considered in the absence of particle transport simulating respiratory tract absorption. The assignment of the compounds to absorption Types F, M or S (fast, moderate and slow), is carried out using the criteria illustrated in Figure 3. Retention below 13% in 30 days, or earlier, would assign the material to Type F (retention above 13% after 30 days would exclude Type F). Retention above 87% in 180 days, or later, would assign the material to Type S (retention below 87% before 180 days would exclude Type S). Retention in the range 13-87% between 30 and 180 days would positively assign the material to Type M.

Absolute values of standard errors for parameters are given in parenthesis. 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 theoretical value.

Dissolution Kinetics and Dissolution Parameters of PHCF Samples

The dissolution of uranium process stream powder samples was carried out over 103 days. The dissolution kinetics of Cameco process stream samples are shown in Figure 4.

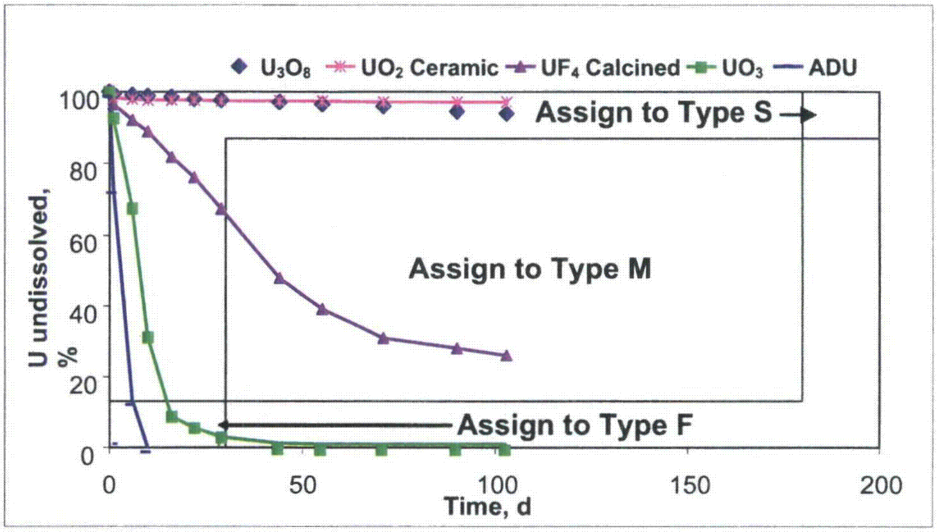

Figure 4 - Dissolution kinetics of Cameco process stream samples

The dissolution parameters for PHCF process stream samples, calculated using one of the equations described above, are given in Table 5.

Based on the ICRP Publication 71 criteria, the PHCF process stream samples could be assigned to the absorption types as shown in Figure 4 and Table 5.

Table 5 - Assignment of PHCF process stream samples to FMS Type

Compound	F_1 (%)	T_1 (d)	F_2 (%)	T_2 (d)	MSD (%)	Absorption Type
ADU	100	2(0.1)	-	-	2.4	F
UO_3	100	6.5(0.5)	-	-	5.6	F
UF_4 calcined	97(8)	44(6)	3(8)	∞	3.6	M
UO_2 ceramic	2.1(0.1)	0.4(0.1)	97.9(0.1)	∞	0.1	S
U_3O_8	0.56(0.1)	0.3(0.4)	99.4(0.1)	∞	0.1	S

Assignment of concentrate samples to absorption types is presented in Table 6.

Table 6 - Assignment of concentrate samples to absorption types

Concentrate Sample / Location/ Experiment year	F_1 (%)	T_1 (d)	F_2 (%)	T_2 (d)	MSD (%)	Absorption Type
Crow 2009, Crow Butte Operation, 2009	78.5(8.6)	0.2(0.1)	21.5(8.6)	2.2(0.3)	0.8	F
KLO 2005, Key Lake Operation Concentrate, Packaging Area, 2005	5.3(0.6)	0.11 (0.06)	94.7(0.6)	∞ (∞)	0.3	S
RLO 2009, Rabbit Lake Operation Concentrate, 2009	93.5(4.2)	7.0(1.9)	6.5(4.2)	∞ (∞)	2.3	F(M)
SRH 2009, Smith Ranch Operation Concentrate, 2009	88.8(4.6)	1.0(0.2)	11.2(4.6)	61.3 (14.6)	2.4	F

Dissolution Kinetics and Dissolution Parameters of Cameco Concentrate Samples

The dissolution kinetics of Cameco concentrate samples are shown in Figure 5.

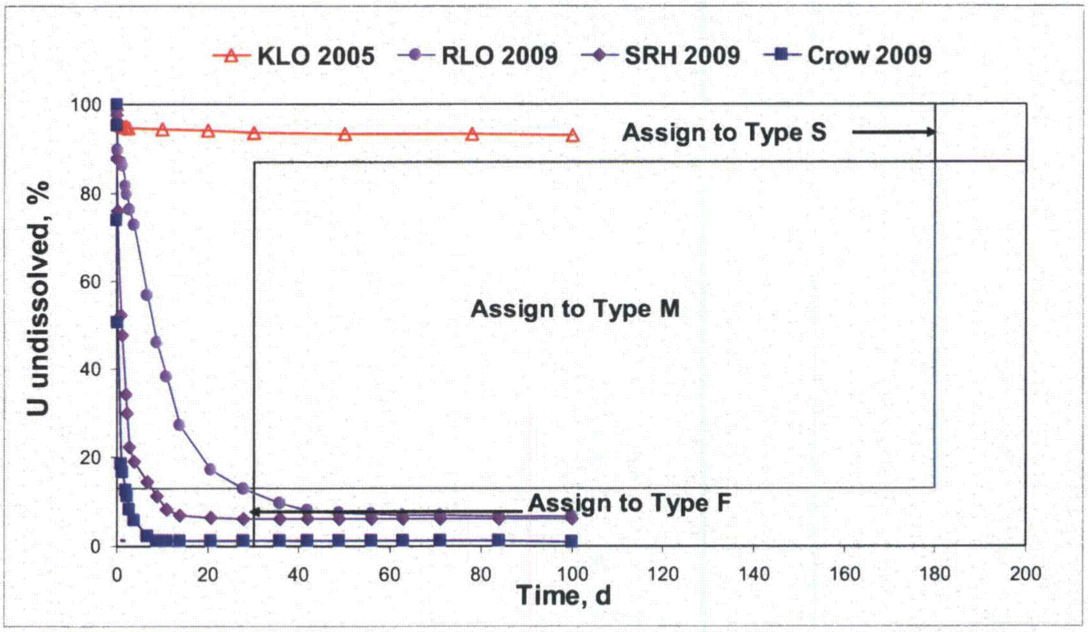

Figure 5 - Dissolution kinetics of concentrate samples

CONCLUSIONS

Dissolution studies of Cameco's site-specific radionuclides were carried out. A new software for calculations of solubility parameters and assignment to FMS absorption types, based on the ICRP Publication 71 requirements, was developed and applied.

ACKNOWLEDGEMENTS

Site personnel at PHCF, CBO, SRH and RLO collected the samples. The authors of this report would like to acknowledge the contributions of all team members. Support of this work by BRR, PHCF and research centre analytical services is gratefully acknowledged.

EFERENCES

1 G. Tairova and M. Ioffe, "Uranium Solubility in Simulated Lung Fluid", Cameco Internal Report No. TD-02-14, 2002.

2 "ICRP Publication 30, Limits for Intakes of Radionuclides by Workers, Part 1", Elsevier, Oxford, UK, 1979, 35-46.

3 "Age-Dependent Doses to Members of the Public from Intake of Radionuclides: Part 4. Inhalation Dose Coefficients", ICRP Publication 71", A report of a Task Group Committee 2 of the International Commission on Radiological Protection, Pergamon, 1995, 393-405.

4 O.R.Moss, "Simulants of Lung Interstitial Fluid", Health Physics, Vol. 36, 1976, 447-448.

5 G.Tairova, "Simulated Lung Fluid Testing. Literature Review", Cameco Interoffice Memo, March 21, 2000, P7665.

6 T.T.Mercer, "On the Role of Particle size in the Dissolution of Lung Fluid", Health Physics, Pergamon Press, Vol. 13, 1967, 1211-1221.

7 "Age-dependent Doses to Members of the Public from Intake of Radionuclides, Part 4: Inhalation Dose Coefficients", ICRP Publication 71. A report of a Task Group Committee 2 of the International Commission on Radiological Protection, Pergamon, 1995, 393-405.