### A PRELIMINARY EVALUATION OF COMMINUTION AND SAMPLING STRATEGIES FOR RADIOACTIVE CEMENTED WASTE

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## ABSTRACT

Lixiviation of Hg, U and Cs contaminants and micro-encapsulation of cemented radioactive waste (CRW) are the two main components of a CRW stabilization research project carried out at Natural Resources Canada in collaboration with Atomic Energy of Canada Limited. Unmolding CRW from the storage pail, its fragmentation into a size range suitable for both processes and the collection of a representative sample are three essential steps for providing optimal material conditions for the two studies. Separation of wires, metals and plastic incorporated into CRW samples is also required. A comminution and sampling strategy was developed to address all those needs. Dust emissions and other health and safety concerns were given full consideration.

Surrogate cemented waste (SCW) was initially used for this comminution study where Cu was used as a substitute for U and Hg. SCW was characterized as a friable material through the measurement of the Bond work index of 7.7 kWh/t. A mineralogical investigation and the calibration of material heterogeneity parameters of the sampling error model showed that Cu, Hg and Cs are finely disseminated in the cement matrix. A sampling strategy was built from the model and successfully validated with radioactive waste. A larger than expected sampling error was observed with U due to the formation of large U solid phases, which were not observed with the Cu tracer.

SCW samples were crushed and ground under different rock fragmentation mechanisms: compression (jaw and cone crushers, rod mill), impact (ball mill), attrition, high voltage disintegration and high pressure water (and liquid nitrogen) jetting. Cryogenic grinding was also tested with the attrition mill. Crushing and grinding technologies were assessed against criteria that were gathered from literature surveys, experiential know-how and discussion with the client and field experts. Water jetting and its liquid nitrogen variant were retained for pail cutting and waste unmolding while attrition milling was selected for fine grinding. Sieving and magnetic separation are among the foreseen technologies to be investigated for metal and plastic rejection. A comminution process flowsheet, to be evaluated and validated at the pilot scale, was designed for the waste site comminution. Water recycling remains the main issue to be addressed. A radioactive waste sampling and comminution laboratory was installed and successfully tested. A statistical material balance algorithm was customized for the lixiviation process for designing sampling protocol and improving accuracy of the contaminants inventory.

### 1. INTRODUCTION

Lixiviation of Hg, U and Cs contaminants and micro-encapsulation of cemented radioactive waste (CRW) are the two main components of a research project carried out at Natural Resources Canada in collaboration with Atomic Energy of Canada Limited. Fragmentation of CRW into a size range suitable for both processes is essential and could have a significant impact on the efficiency of both processes as a result of the micro-cracks generation and the fineness homogeneity of the size distribution of the waste after grinding. Sampling is another important issue considering the rather small sample weight (2 -10 g) required for the lixiviation exploratory testwork and the heterogeneity distribution of the contaminants within the waste matrix. Sampling and comminution are intimately related in the feed preparation process and they are therefore addressed within the same study. Health and safety issues mainly related to radioactive and toxic dust emissions, and the presence of wires, metals and plastics in CRW are two important concerns to be addressed within the study.

The objectives of the study were to gather basic information on the surrogate cemented waste (SCW) properties, to develop a sampling strategy, to characterize and document the SCW response to different grinding mechanisms, to identify the most suitable comminution strategy and to determine the possibility of these technologies to be adapted to the waste-site comminution of the radioactive cemented waste which is contained in steel pails of about 20 kg each. The deliverables were the installation of a radioactive waste sampling and comminution laboratory and the development of a process flowsheet to be evaluated at the pilot scale in a future phase. Surrogate cemented waste (SCW), where Cu was used as a substitute for U and Hg, was initially used in the study as it is safer to use and does not affect the mechanical properties and the comminution response of the waste. The sampling model initially developed with SCW was later validated with radioactive waste. The report presents briefly the composition of the radioactive cemented waste, the methodology and the achieved results.

## 2. CEMENTED WASTE COMPOSITION

The compositions of both the surrogate radioactive waste (SRW) and non-radioactive surrogate cemented waste (SCW) are presented in detail by Fiset and co-authors [14]. They both consist of 15.5 kg of general usage cement mixed with 4.5 litres of an aqueous solution containing the different contaminants. In the case of the surrogate waste, Hg and U were replaced by Cu with similar concentration. A summary of the metal constituents of the radioactive solution is presented in Table 1.

Constituent	Concentration	Chemical formula	Formula weight	Amount required for 4.5L	Assay
	mol/L		g/mol	g	ppm
Aluminium	0.98	Al(NO <sub>3</sub> ) <sub>3</sub> .9H <sub>2</sub> O	375.13	1650	5934
Uranium	0.02	$UO_2(NO_3)_3.6H_2O$	502.13	45.2	1071
Mercury	0.033	$Hg(NO_3)_2.H_2O$	342.61	50.9	1490
Rubidium	7.53E-05	RbNO <sub>3</sub>	147.47	0.05	1.45
Cesium	2.19E-04	CsNO <sub>3</sub>	194.91	0.192	6.55
Strontium	2.96E-04	$Sr(NO_3)_2$	211.63	0.281	5.82
Barium	2.29E-04	$Ba(NO_3)_2$	261.35	0.269	7.07
Rubidium	3.02E-04	RuNO(NO <sub>3</sub> ) <sub>3</sub> as		9.16	6.87
		1.5% solution			
Lanthanum	1.44E-04	$La(NO_3)_2.6H_2O$	433.02	0.281	4.51
Cerium	3.89E-04	$Ce(NO_3)_3.6H_2O$	434.23	0.76	12.3
Praseodymium	1.01E-04	$Pr(NO_3)_3.6H_2O$	435.03	0.198	3.21
Neodymium	2.84E-04	$Nd(NO_3)_3.6H_2O$	438.35	0.561	9.23
Samarium	3.40E-05	$Sm(NO_3)_3.6H_2O$	444.46	0.068	1.15
Europium	4.38E-06	$Eu(NO_3)_3.6H_2O$	446.07	0.009	0.17
Gadolinium	1.41E-06	$Gd(NO_3)_3.6H_2O$	451.36	0.003	0.05
yttrium	1.28E-04	$Y(NO_3)_3.6H_2O$	383.01	0.22	2.55
Iron	1.00E-03	Fe(NO <sub>3</sub> ) <sub>3</sub> .9H <sub>2</sub> O	404	1.82	12.6
Nickel	1.80E-04	Ni(NO <sub>3</sub> ) <sub>2</sub> .6H <sub>2</sub> O	290.81	0.23	2.32
Chromium	3.30E-04	Cr(NO <sub>3</sub> ) <sub>3</sub> .9H <sub>2</sub> O	400.15	0.59	3.83
		HNO <sub>3</sub> (16M)		100	

Table 1.	Summary	of metal	constituents	of aqueous	solution [14]
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# 3. METHODOLOGY

## 3.1 Surrogate Cemented Waste (SCW) Characterization

## 3.1.1 Contaminants heterogeneity distribution

For the initial part of the study, the surrogate cemented waste (SCW) was disengaged from the pail simply by hammering the side and the bottom of the container. Figure 1 shows an out-of-the pail sample.

It was found that the SCW sample was friable and could be disintegrated by hand. A size-bysize analysis of the material was performed before fragmentation to evaluate the distribution of the contaminants as a function of particle size. At the microscopic scale, the heterogeneity distribution of Cu, Cs, U and Hg was studied by SEM microscopy and through the calibration of the sampling model (Eq. 1) initially proposed by P. Gy [18] and modified by D. François-Bongarcon [16], [27].



Figure 1. Surrogate cemented waste sample as removed from the pail.

The model (Eq. 1) evaluates the sampling error as a function of the 95% passing size (d), the mass of the lot ( $M_L$ ), the mass of the collected sample ( $M_s$ ) and two parameters K and  $\alpha$ , that characterize the heterogeneity of the material. The sampling tree methodology originally proposed by François-Bongarçon [15] and recommended by a number of authors [1] and [26] was selected for calibrating the material heterogeneity constants. Other techniques published in the literature [13], [25], [31] were also considered. The contaminants heterogeneity parameters were initially calibrated with the non-radioactive surrogate material and the resulting model was later validated with the radioactive surrogate waste [7], [8].

$$\sigma^{2}(FE) = Kd^{\alpha} \left[ \frac{1}{M_{s}} - \frac{1}{M_{L}} \right] \qquad (1)$$

#### 3.1.2 Hardness index

The hardness of the material was characterized through the measurement of the Bond work index [9], which is widely used in the mining industry for ore hardness evaluation, and sizing of comminution equipments [31]. The Bond work index gives the hardness parameter (Wi) that is used (Eq. 2) to evaluate the energy required (W) to fragment a material from a feed size of  $F_{80}$  to a product size of  $P_{80}$  [9].  $F_{80}$  is the size of a sieve in micrometers through which 80% of the feed material passes. Similarly,  $P_{80}$  is the size in micrometers through which 80% of the product passes.

$$W = \frac{10^* W_i}{\sqrt{P_{80}}} - \frac{10^* W_i}{\sqrt{F_{80}}}$$
(2)

### 3.2 Design and assessment of a sampling strategy

Equation 3, which is another form of Equation 1, allows to evaluate the relative variance of the minimal sampling error as a function of the material heterogeneity constants ( $K,\alpha$ ), its nominal top size (d) and the sample weight (Ms).

$$\ln(\sigma^{2}(FE)) = (-1)\ln(Ms) + [\alpha \ln d + \ln(K)]$$
(3)

On a log-log graph showing the variance of the sampling error as a function of the sample weight (Figure 2), Equation 3 generates for a given material heterogeneity (K, $\alpha$ ) a series of parallel lines having a slope of -1, corresponding to different nominal top-sizes. This graph is known as the sampling nomogram [1], [15], [26]. It is mainly used to define the sample mass splitting and stepwise size reduction scheme that should be followed to achieve a given accuracy in measuring concentration of the elements of interest. It consists essentially of the evaluation of the sampling error at each mass and size reduction steps between the initial sample taken from the lot and 1 g sample used for chemical assays.

A comparison of two sampling protocols is provided as an example in Figure 2. In the 1<sup>st</sup> sampling protocol, an initial sample of 12 kg was collected at 1 cm (a), crushed to 1 mm (a-b), sub-sampled to 1 kg, (b-c), pulverized to 75  $\mu$ m (c-d) and finally sub-sampled to 70 g for chemical assay (d-e). The total error of such a sampling process is about 16% ( $\sigma^2 = \sigma^2(a) + (\sigma^2(c) - \sigma^2(b)) + (\sigma^2(c) - \sigma^2(d))$ ). In the second sampling protocol (dashed line) the primary sample (a) is pulverized immediately to 75  $\mu$ m (a-f) before being sub-sampled to 70 g (f-e) for chemical assay. The total sampling error is reduced to 13% ( $\sigma^2 = \sigma^2(a) + (\sigma^2(c) - \sigma^2(f))$ ) at the cost of additional pulverizing work.

A sampling methodology was derived using the concept of the sampling nomogram. The flowchart of the sampling methodology is shown in Figure 3. It is an iterative process where a sampling nomogram, computed from the minimal sampling error model and the SCW heterogeneity parameters, is used to estimate the sampling errors for the initial laboratory test design specifications and the sampling protocol proposed to collect these samples. The accuracy of the metallurgical results is then evaluated as a function of the sampling and analytical errors through a lixiviation material balance simulation. If the accuracy is not within the acceptable limits, then an alternative sampling protocol (for example sampling after grinding rather than sampling after crushing only) is evaluated until the target accuracy is obtained. A range of sample weights to be collected at certain number of nominal top sizes was then established and the comminution and sampling equipment type and operating ranges were selected accordingly.

The material balance algorithm BILMAT, which is used for that purpose in the mining industry, was modified for the lixiviation process. Further details about material balance simulations are given elsewhere [2], [5], [20].



Figure 2. Example of sampling nomogram.

Due to delays in developing an assay method for Cs, Hg and U in the radioactive cemented waste, the validation of the sampling model was limited in measuring the representativeness of a number head samples produced for the lixiviation testwork. Further validation testwork based on a statistical material balance analysis of the hydro metallurgical testwork will take place in a near future.

#### **3.3** Assessment of comminution technologies

In conventional comminution equipments, material fragmentation takes place through three fundamental mechanisms, which are compression, impact and attrition [39]. Most of the industrial and laboratory comminution equipment uses a combination of these three mechanisms for rock breakage; however, they are usually classified, based on their predominant breakage mechanism. The SCW response to each breakage mechanism was evaluated using the equipment listed in Table 2. Expected main characteristics of the ground materials produced by each of the selected technologies are also presented in Table 2. Both wet and dry grinding modes were tested. Autogenous and semi-autogenous grinding tests were not justified because the cemented waste is very friable and thus lacks the significant percentage of harder material required for use as a grinding media in such methods [29].



Figure 3. Flowchart of the sampling design methodology.

A less conventional approach, cryogenic grinding [11], [24] was also tested with the attrition mill. In this mode, the material is cooled down to its brittle temperature either by using liquid nitrogen or carbon dioxide [4] prior to subjecting it to fragmentation. Hammer mill or attrition mill are commonly used in cryogenic grinding [10], [35]. This technique is used for recycling tires and in grinding elastic and heat sensitive materials [19]. For the attrition mill, the cooling system is integrated into the grinding chamber for more efficiency [38]. The energy required for the impact breakage of steel as a function of temperature [30] suggests that cryogenic grinding could be an option for steel pail fragmentation. One of the main advantages of cryogenic grinding in this project would be its ability to grind soft material such as plastic and the resulting opportunity of dismantling the pail-bag-cemented sample in a confined chamber. Cryogenic grinding has the advantage of dust control without the problematic generation of radioactive water. The absence of water in the grinding chamber would additionally provide a better control on Cs lixiviation as desired for the hydrometallurgy tests.

Two other less conventional fragmentation techniques where there is no mechanical contact between the equipment and the waste were also assessed: the High Pressure Water Jetting and the High Voltage Pulse Power technologies [3], [12]. Their main foreseen advantage in this project are 1) dust-free material unmolding potential; 2) robustness of the equipment in the presence of steel wire and other metals that cannot be ground or crushed mechanically; 3) the absence of grinding media that may generate additional contaminants (Fe, Cr, Ni).

Fragmentation mechanism	Grinding/Crushing technique	Product size distribution main characteristics
Compression	Jaw crusher	Two population - coarse and fine size fraction
	Cone crusher	More homogeneous in comparison to jaw crusher
	Rod mill	Reduced proportion of very fine size fraction
Impact	Ball mill	More homogeneous size distribution
Attrition	Attrition mill	Finer size distribution
	Electro-dynamic	Preservation of natural particle size
	Electro-hydraulic	distribution
Water jetting	Forced Pulse	Coarse fragment and large block size
	Ultra-high pressure	
	Liquid nitrogen	

Table 2. Investigated crushing and grinding techniques

The high voltage fragmentation breaks the material either by tension through a high voltage discharged into the solid (electro-dynamic mode) or by compression via a pulse resulting from a high voltage discharge into the liquid (electro-hydraulic mode). The selFrag-XYZ system [33] was proposed by the manufacturer as a possible solution for the waste-site comminution of the radioactive concrete. This system offers the possibility to manipulate the discharge electrode in three perpendicular directions over a vessel, in which up to seven pails containing cemented waste could be processed. Further testing would be required of this system for pail waste unmolding and primary grinding. The coarse product would eventually feed a secondary grinding process.

High pressure water jet is a mature technology that has been used for more than thirty years in cleaning applications, recycling of concrete, hydro-demolition, metal cutting, mining, comminution, etc. [23], [34]. This technology breaks the material through the impact of the water jet that penetrates pre-existing flaws, which grow as the fracture process is initiated and eventually intersect and form fragments [28]. The basic parts of a high pressure system include a source of water, a pump, a mobile arm, a hose and a lance to which the nozzle is attached [17]. In general, systems operating at pressures below 30 k-psi are known simply as high pressure water jet, whereas systems operating at 30 to 90 k-psi are known as ultra-high pressure water jet [22].

A few variants of the high pressure water jetting that were tested for SCW unmolding and grinding are: 1) the standard High Pressure Water Jet, 2) the Force Pulse Water Jet (FPWJ), 3) the Ultra-High Pressure Water Jet, and 4) the Liquid Nitrogen Blasting technology. The liquid nitrogen blasting technology is a variant of the high pressure water technology in which the water is replaced by liquid nitrogen. The FPWJ is a relatively newer technology patented in 1992 by Mohan Vijay. In this technique pulsing of the jet using an ultrasonic nozzle device is used to increase the impact on the target by a factor of 6-12 depending on the pump- operating

pressure [36], [37]. The basic manufacturer descriptions of the FPWJ and Nitrogen Blasting technology are presented on their respective web sites.

Many applications of the high pressure water jet technology and different equipment configurations are given by the manufacturers and also in textbooks [27], [34]. It is worthwhile mentioning that this equipment has already been used in removal of radioactive material. The U.S. department of Energy is using high pressure water for cleaning the 149 Hanford aging tanks from its radioactive solids and transfer the solids into 28 newer and more secure double-shell tanks [21]. A robotized system called the Mobile Arm Retrieval System (MARS) equipped with a high pressure nozzle that spray 20-30 gallons a minute at a pressure of 5 k psi was later developed by this organization. It is now being tested on non-radioactive material. The MARS system is remotely controlled using a combination of camera, joysticks, pushbuttons and switches. A second version combining the water jet to a vacuum system for material removing is being developed.

The main assessment criteria that have been addressed through the testwork and the many consultations with the client and technology suppliers are: 1) waste engineering capabilities; 2) equipment scale-up and operating; 3) maintenance and reliability; 4) health and safety issues including radioactive dust emission; and 5) waste water generation and recycling. For safety purposes, the required testwork was performed with a surrogate cemented material with no radioactive compounds but with similar mechanical properties.

## 4. RESULTS AND DISCUSSION

### 4.1 Surrogate Cemented Waste Characteristics

A visual observation and manipulation of many SCW pail samples did not show any significant difference among them in terms of hardness and size distribution measured before any mechanical fragmentation. The size distribution analysis of the cemented waste (Figure 4) shows a significant percentage of fine material, smaller than 45  $\mu$ m, in the sample. This fine material is the range of cement powder and is believed to be un-hydrated cement. Figure 4 also gives the Cs assay in each size fraction, and it shows that the fine material contains much less Cs than the larger blocks.

A number of the large blocks could easily be broken by hand and this high friability of the waste was confirmed in laboratory through the measurement of a Bond work index of 7.7 kWh/t. As a comparison, clay is characterized by a work index of 7.



Figure 4. Size distribution of SCW and Cs contaminant by size fraction before crushing.

The heterogeneity distribution of the contaminants within the cemented matrix is observed visually after unmolding the waste from its disposal pail (Figure 1). This heterogeneity is due to the limited mixing of cement and solution in the pail. A mineralogical investigation and the calibration of the material heterogeneity parameters of the sampling model show that Cu and Cs are finely disseminated in the cement matrix. They appear to be in concentration proportional to the hydration water, and the amount of hydrated cement in the pail is heterogeneous. There is more hydrated cement in the bottom of the pails than in the top of the pails. The Cu and Cs do not react with cement to produce discrete phases and they rather remain in the hydration water at the size of their atomic diameter, which is for example about 2.6 Å for Cu. The result of the contaminant distribution analysis by size fraction (Figure 4) is coherent with this observation; it shows a lower concentration of Cu (tracer) and Cs in the minus 45  $\mu$ m size fraction, which is most probably explained by a lower hydration of the cement in this size fraction.

The mineralogical analysis of the radioactive waste indicated a different behaviour for Hg and U. Ca-U solid phase was observed as grains of up to 20  $\mu$ m and also as thin layers as large as 1 mm by 5 mm [14]. Solid phases were also observed for Hg but their sizes were of a few microns only (3 to 1  $\mu$ m). This more heterogeneous distribution of U and Hg was not predicted by Cu, which was used as a tracer for both contaminants.

## 4.2 Sampling strategy

A sampling nomogram was computed for Cu and Cs after calibration of the sampling model. It is presented for Cu in Figure 5; information for Cs was very similar. The nomogram shows that crushing the 20 kg waste sample at a nominal top size of about 2 mm overcomes the macro heterogeneity zones of the pail for samples larger than 100 g. Smaller samples of 10 g and less should be collected at a nominal top size of at least 400  $\mu$ m in order to keep the variance of sampling in the same range. It was verified by material balance simulation that such a sampling error was acceptable for providing representative samples for the lixiviation testwork. A sampling protocol was developed accordingly. It consists of crushing (2 mm) and splitting the 20 kg content of the pail into sub-samples of 1000 g and grinding a sub-sample to a nominal top size of about 40  $\mu$ m before collecting smaller samples in the range of 6 g each. Rotary splitters were used for samples splitting. Riffler could also be used but it is important to mention that the riffle chute openings should be at least 20 mm in width when the sampling is done after crushing.

As indicated, the validation of the sampling strategy was limited to a number of surrogate radioactive cemented waste head samples that were produced for the lixiviation testwork. The analysis consists of comparing the measured average assay of Cs, Hg and U, their standard deviation and the calculated sampling error to their expected values (Table 3). The expected values are derived from the 4.5 litres of solution (Table 1) used to mix with 15.5 kg of cement. The sampling error was calculated based on a measurement error of 5.0% for Cs and 2.5% for Hg and U, which were estimated in a previous study [7]. It is concluded from the data in Table 3 that the sampling error is however larger than predicted for U and this is more probably due to the presence of larger Ca-U phases that were observed by the mineralogical characterization and not predicted by the Cu tracer.



Figure 5. Cu sampling nomogram.

For all of the contaminants it is also observed in Table 3 that the measured assays are more than three standard deviations away from the expected assay values. For Cs, such a difference is not a concern as it most probably results from the initial concentration of Cs in the cement powder. For Hg and U it was verified that these differences are more likely due to the fact that the expected values were calculated for wet-base samples, whereas the assays are measured on drybase samples, which are reduced in weight by about 10%.

	Measured values			Expected values			
	Assay	Standard deviation	Sampling error	Assay	Standard deviation	Sampling error	
	(ppm)	(%)	(%)	(ppm)	(%)	(%)	
Hg	1669	2.4	< 0.3	1490	2.5	0.3	
Cs	8	4.4	< 0.3	6.6	5.0	0.3	
U	1172	3.1	1.9	1071	2.5	0.3	

### Table 3. Representativeness of the lixiviation head samples

## 4.3 Comminution process

The waste engineering capabilities assessment has shown that none of the technologies fulfill all the on-site radioactive waste comminution needs. A combination of technologies is therefore needed for the comminution of the cemented waste in a safe environment. Attrition milling is the closest to autonomy and will constitute the central grinding equipment of the comminution process flowsheet (Figure 6) that was designed based on the technologies assessment. It was demonstrated that the attrition mill can grind the waste from blocks of about 15 cm diameter down to 20  $\mu$ m, which is definitely fine enough for feeding the lixiviation process. Such a fine grind was obtained in wet, dry and cryogenic mode. This technology was also successfully used to grind radioactive samples using the Union Process laboratory unit, model 01-HD. Our experience in grinding the waste with this technology suggests however that it could be difficult to control the product size at size larger than a few millimetres, which could be mandatory if micro-encapsulation alone without pre-lixiviation is retained by the client for waste treatment. The use of larger balls in the grinding media could be an option if this ever becomes a requirement.

Processes required for preparing the feed to the attrition mill include the pail opening and/or cutting, the unmolding of the concrete waste from its containing pail, the primary crushing of the waste and the separation of plastic and steel pail fragments and other metallic pieces (Figure 6). Sieving, magnetic and gravity separation are among the foreseen technologies to be investigated for metal and plastic rejection. Testwork realized has shown that high pressure water jet technology, either in pulsed or in ultra high pressure mode, is the most promising technology for the pail cutting, waste unmolding and the primary crushing of the waste. Its main advantages against high voltage discharge fragmentation are its smaller but still important consumption of water; the reduced treatment required for water recycling; and its simplicity of operation.

A filtration system removing particles larger than 5  $\mu$ m is needed for water recycling as the high pressure water jet generates between 30 and 126 litres of dirty water per pail, which is more than the 24 and 6 litres per pail required respectively for the lixiviation and the micro-encapsulation processes. Recycling of radioactive water complicates the maintenance requirements as all the parts of the high pressure system (pump, hose and nozzle) would become contaminated. Potential explosion of a high pressure hose filled with radioactive water should also be considered and its risk management properly planned. The nozzle is the most important maintenance issue as it needs replacement after 40 to 500 hours of operation. The nozzle would also be the only piece of equipment in contact with radioactive material if water recycling can be avoided.

If the water jetting process is operated in batch mode and enough time is given between each pail for water decantation and filtration, then the total amount of water required for processing a batch of seven pails would be ceiled to 126 or 60 litres per batch depending on the technology used. Such a volume of water would be actually less than the 175 litres of water required for the lixiviation of seven pails and this would address the client concern about the accumulation and mass balance of the contaminated water. If lixiviation is not retained in the waste treatment process, there will still be however, accumulation of contaminated water as the micro-encapsulation should not use more than 42 litres of water per batch of seven pails.

The liquid nitrogen variant of the high pressure water jet technology could be an option to water recycling but the entrainment of the radioactive dust in the nitrogen gas and the amount of abrasive required for cutting the pail needs to be further investigated. Another option is to cut the pail around the waste instead of unmolding the waste from the pail. The use of an ultra-high pressure system in which the flow of water is reduced from 36 to 3.4 litres/min could also be part of the solution. All those options need to be further investigated.



Figure 6. Proposed comminution process flowsheet.

# 5. CONCLUSIONS

Surrogate cemented waste (SCW) was initially used for this comminution study where Cu was used as a substitute for U and Hg. SCW was characterized as a friable material through the measurement of Bond work index of 7.7 kWh/t. A mineralogical investigation and the calibration of the material heterogeneity parameters showed that Cu, Hg and Cs are rather finely disseminated in the cement matrix. A sampling strategy was built from the model and successfully validated with radioactive waste. A larger than expected sampling error was observed with U due to the formation of larger Ca-U solid phases, which were not observed with the Cu tracer.

SCW samples were crushed and ground under different rock fragmentation mechanisms: compression (jaw and cone crushers, rod mill), impact (ball mill), attrition, high voltage disintegration and high pressure water (and liquid nitrogen) jetting. Cryogenic grinding was tested in addition to wet and dry grinding with the attrition mill. Crushing and grinding technologies were assessed against criteria that were gathered from literature surveys, experiential know-how and discussions with the client and field experts. Water jetting and its liquid nitrogen variant were selected for pail cutting and waste unmolding while attrition milling was retained for fine grinding. Sieving and magnetic separation are among the foreseen technologies to be investigated for metal and plastic rejection. A comminution process flowsheet was designed for the waste site comminution based on the technologies assessment results. Water recycling remains the main issue to be addressed. A radioactive waste sampling and comminution laboratory was installed accordingly and successfully tested. Experiments were designed to evaluate and validate the proposed flowsheet at the pilot scale.

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