EVALUATION OF THIN-FILM EVAPORATION FOR DECONTAMINATION AND IMMOBILIZATION OF AQUEOUS NUCLEAR WASTE

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ABSTRACT

In the early 1980's, AECL, at the Chalk River Laboratory (CRL) site, built a Waste Treatment Centre (WTC) for managing low level solid and aqueous liquid wastes. The objective was to demonstrate processes for converting Canadian Deuterium Uranium (CANDU) wastes to a form suitable for disposal while meeting or exceeding current environmental regulations.

At present, two liquid waste streams are being treated at the Waste Treatment Centre. The liquid waste streams are volume reduced by a combination of continuous crossflow microfiltration (MF), spiral wound reverse osmosis (SWRO), and tubular reverse osmosis (TRO) membrane technologies [1]. The concentrate produced from the TRO system and the volume-reduced MF backwash solutions are evaporated while simultaneously adding bitumen in a thin-film evaporator. A water-free product of chemical and radiochemical salts and bitumen is removed in 200 L galvanized steel drums for storage and eventual disposal in the CRL Waste Management Area.

The feed stream to the thin-film evaporator typically has a β/γ activity of about 1 - 3 μCi/mL. This intermediate-level radioactive stream is concentrated by a factor of about 10, while simultaneously being immobilized. The radiation field of product drums on contact typically has a value of 0.5 to 3 R/h depending upon the feed concentration of radioactivity to the evaporator. The total solids content in the 200 L drum ranges from 25% to 35%. Encapsulated in the bitumen matrix are a variety of non-radiochemical salts (including sodium phosphate, sodium sulphate, and sodium carbonate) which comprise the bulk of the total solids in the product drum. The drum contains less than 1% of free water.

The paper will discuss the volume reduction capability of the plant, with an emphasis on the immobilization of the aqueous waste with bitumen in a thin-film evaporator. Operations experience gained from over 200 campaigns is documented in the paper.

INTRODUCTION

In 1975, the need for a Waste Treatment Centre (WTC) at Chalk River Laboratories (CRL) was identified. The WTC was built to demonstrate systems capable of converting liquid wastes to a stable and leach-resistant form. Current operations in the facility involve the treatment of the low-level CRL liquid waste using a 3-stage membrane plant to accomplish most of the volume reduction (comprised of microfiltration, spiral wound reverse osmosis, and tubular reverse osmosis unit operations). Further volume reduction is achieved with a thin-film evaporator (without bitumen addition), and another thin-film evaporator whose primary function is for bitumen immobilization.

Fig. 1

Integrated Radwaste Processing Plant at AECL

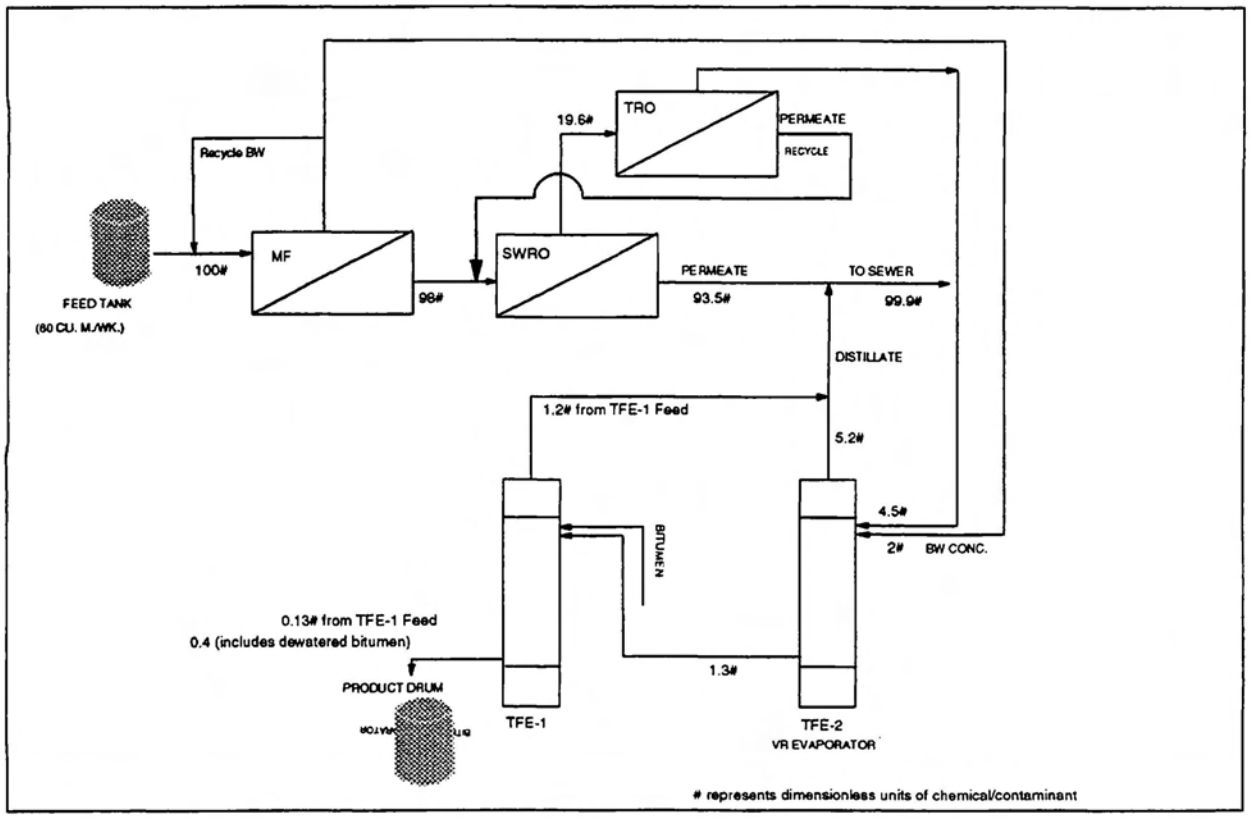


Figure 1 shows a flowsheet and volumetric balance for the primary unit operations at the WTC. For a basis of 100 units of liquid waste (or 100# in the figure), the quantities of liquid at various points in the plant are indicated. A total volume of 93.5# is released by the 2-stage reverse osmosis plant (per 100# treated), while a combined 99.9# is released from both the reverse osmosis and thin-film evaporator operations. A total volume of 0.4# (including emulsified bitumen volume) is the secondary waste product from the thin-film evaporator used for bituminizing (TFE-1). This results in an overall volume reduction factor of 250 for the plant, based on feed liquid volume to immobilized secondary waste volume, in the product drum (that is, 100 ÷ 0.4).

The TFE-1 unit is used to immobilize liquid waste concentrates with bitumen. The other (TFE-2) is used to volume reduce microfiltration backwash concentrates and tubular reverse osmosis retentate prior to immobilization. The bituminized product from TFE-1 is collected in 55 US gallon (210 L) galvanized steel drums. A full drum weighs approximately 240 kg. The drums are capped, transferred to a shielded container and then shipped to the site waste management area for storage and eventual disposal. The evaporation/immobilization process is carried out remotely in a shielded facility that is designed to handle product drums with contact gamma radiation fields as high as 10 R/h. In practice, however, the radiation fields are maintained below 5 R/h.

At present, two radioactive CRL liquid waste streams are being processed by the thin-film evaporator used for bitumen immobilization (TFE-1). One stream originates from the central Decontamination Centre (DC). The other, an Active Drain (AD) stream, is produced from a large and diverse number of research laboratories and radioisotope production facilities. The two waste streams,

totaling about 3000 m^3 per year are volume reduced by a factor of 80 prior to immobilization by three membrane systems and thin-film evaporation.

TECHNOLOGY DESCRIPTION

Membrane Plant

A flowsheet of the integrated plant for volume reduction and immobilization of aqueous radwaste is shown in Fig. 1. Liquid waste feed to the WTC is sampled and analysed for pH, conductivity, α and gross β/γ activity, tritium (^3H), turbidity, total solids, gamma-emitting radionuclides, and various non-radioactive chemical species at the source tanks. Waste is then sent to a 45 m^3 feed tank in the WTC and it is pH adjusted to the alkaline region (8 - 10); an adequate conditioning time of 12 hours is allowed to maximize precipitation of metals, prior to treatment with MF for the removal of suspended solids.

During microfiltration (MF), concentrate is recycled; filtrate is continually removed from each bank of filtration modules at about 25 L/min, and is directed to the spiral wound reverse osmosis (SWRO) feed tanks. When the backwash (concentrate) solution exceeds 10 g/L, it is sent to the volume reduction evaporator, TFE-2. The MF filtrate, free of suspended solids, enters the SWRO feed tank at a rate equal to the permeation rate of the SWRO. The concentrate from the SWRO membrane process is bled to the TRO feed tanks.

The TRO membrane system operates in a batch process mode, where the final tank volume is reduced to approximately 10 to 15% of the initial feed volume. The concentrate, containing about 50 to 80 g/L of dissolved solids, is transferred to one of three 7100 L tanks in the active tank room, to await further volume reduction by evaporation with TFE-2.

Thin-film Evaporator Process Description

Each of the two thin-film evaporators has a 1 m^2 heated surface, and is operated with superheated steam at about 720 kPa applied to the jacket. Waste feed containing between 5-15% total solids is introduced at a rate of about 1 L/min, along with bitumen emulsion flowing at about 0.3-0.5 L/min. The emulsified bitumen and the waste are introduced via a distribution ring to the top of the evaporator and fall along the heated surface. The evaporator has internal rotating blades, which sweep within 1 mm of the vertical heated surface. The blades in TFE-1 rotate at 900 rpm.

The evaporator does not operate full of product; the liquid or slurry forms a thin film or annular ring of product from the feed nozzle to the product outlet nozzle. Holdup or liquid inventory in a thin-film evaporator is very low, typically about 5 kg of material per square metre. A thin-film evaporator, which is an inherently low-pressure-drop device, has mechanical turbulence and, therefore, good heat transfer properties over a wide range of viscosities [2].

PERFORMANCE OF INTEGRATED PLANT FOR REMOVAL OF CONTAMINANTS

The removal efficiencies for phosphate and some of the key radioactive substances are shown in Table 1 for the integrated plant.

Table 1
Removal Efficiencies of MF/SWRO and Integrated Plant

FEED [1]	PERMEATE [2]	REMOVAL	DISTILLATE	PLANT
Ci	Ci	% [3]	Ci [4]	% [5]

ALPHA	0.30	0.0016	99.45	0.0003	99.36
BETA	61.08	1.0127	98.34	0.0114	98.32
Co-60	4.49	0.0205	99.54	0.0008	99.52
I-131	1.67	0.0011	99.94	0.0000	99.94
Cs-137	11.54	0.3665	96.82	0.0197	96.65
Ce-144	4.70	0.0089	99.81	0.0005	99.80
Sr-90	25.41	0.0530	99.79	0.0076	99.76
PO4 (kg)	340.00	1.84	99.46	0.62	99.28

Notes:

1. Radioactivity in feed to integrated plant, in curies.
2. Radioactivity in permeate from SWRO plant, in curies.
3. Removal efficiency with SWRO membrane plant.
4. Radioactivity in distillate stream from evaporators TFE-1 and TFE-2, in curies.
5. Removal efficiency with integrated plant, including membranes and evaporators.

The removal efficiency of the two-step membrane plant employing MF and SWRO is indicated in the forth column as removal percent. The removal efficiency of the MF/SWRO system for gross α is 99.45%. In comparison, 98.32% of the gross β/γ emitters (excluding tritium) is removed. The removal efficiency of gross β/γ is lower in the membrane plant due to the relatively low removal efficiency of the cesium isotopes, including Cs-137. Only 96.82% of the Cs-137 is removed by the MF/SWRO system. In comparison, removal efficiencies of the other dominant radionuclides are: 99.54% for Co-60, 99.94% for I-131, 99.81% for Ce-144, and 99.79% for Sr-90. A method to improve the efficiency of the MF/SWRO system for cesium removal would be desirable from a processing perspective.

Phosphate is one of the chemicals in the AECL effluent which is subject to Federal Discharge guidelines [3]. As a result, close attention is paid to the performance of the plant for phosphate removal. For the MF/SWRO system the removal efficiency was 99.46%. The effluent quality consistently met the Canadian Discharge guidelines.

The removal efficiency of the integrated plant, including the two thin-film evaporators, is indicated in the last column of Table 1 as plant per cent. The overall plant efficiency is lower than for the membrane plant alone, since there is an additional mass of contaminants lost with the distillate stream. The overall removal efficiencies for the integrated plant decrease only slightly, since the distillate stream represents a small fraction of the effluent (about 150 m^3 per year) from the integrated plant. It should be noted that the removal efficiencies of the evaporators themselves usually exceeds 99.9% for all contaminants, including Cs-137 [4].

BACKGROUND ON SOLIDIFICATION AND IMMOBILIZATION TECHNOLOGY

The purpose of solidification technology is to produce solids that are nonhazardous, or less hazardous than the original waste components. The degree of hazard for these types of materials or systems is usually determined by leach tests. Solidification technologies not only solidify the waste by chemical means, but also insolubilize, immobilize, encapsulate, destroy, sorb, or otherwise interact with selected waste components.

The terms stabilization and solidification are used interchangeably in the literature. Normally "solidification" is reserved for liquids, although

references have also been made to solids [5]. "Immobilization" usually refers to encapsulation of waste such that the contaminants are immobile. It usually involves adding materials that will ensure that the hazardous constituents are maintained in their least mobile toxic form. Solidification implies that the beneficial results of treatment are obtained primarily, but not necessarily exclusively, through the production of a solid block of waste material with high structural integrity -a product often referred to as a monolith. The contaminants do not necessarily chemically react with reagents. Instead, they are mechanically locked within the solidified product. This is known as microencapsulation. Contaminant loss is minimized by decreasing the surface area exposed to the environment or isolating the contaminants by microencapsulating the waste particles.

Types of Binders Available

The currently applied materials for solidification of wastes are: 1) hydraulic cements, pozzolans, and gypsum, which harden by reacting with water, 2) thermoplastic bitumen, polyethylene, and sulphur, which melt and freeze encapsulating the waste solids, and 3) polyesters, epoxies, poly-urethanes, and urea-formaldehydes whose monomers react to form cross-linked polymer chains around the waste. The most widely used of these are the hydraulic based binders. The dominant inorganic (hydraulic-based) binding materials at the present time include either Portland cement alone (or with minor additives), cement or lime kiln dust, Portland cement-flyash, lime-flyash (or other pozzolan), lime-based, and Portland cement-sodium silicate. Portland cement has been the most widely used inorganic ingredient in terms of diversity of use, especially when combined with sodium silicate or flyash [5].

Rationale for Selection of Bitumen Matrix at AECL

Solidification of wastes in bitumen has been tested extensively in Europe, and has been applied more recently at Virginia Power in the United States [6]. Experience has demonstrated that bitumen is also suited to most streams generated by nuclear power plants and by industry. The stability of bitumen with respect to radiation is a property of primary importance when bitumen is considered as a waste matrix material. The main factors influencing the radiation stability of bitumen are the dose-rate and the total absorbed dose. A total absorbed dose limit of 10^8 to 10^9 rad is commonly reported for the radiation resistance of bitumen-waste forms [7].

Bitumen generally provides superior leach resistance in comparison with cement, which could be important for certain species and disposal scenarios. One of the potentially more significant advantages provided by bituminization processes is volumetric efficiency, which is particularly important if waste must be stored for extended periods before disposal. Cement solidification can result in as much as a 100% increase in volume, whereas, depending on waste stream concentrations and the type of equipment used, bituminization can provide volume reduction factors of 5 or more, because associated water is driven off during the solidification process.

Description Of Bitumen Used For Immobilization

Bitumen is the name given to a wide range of hydrocarbons with high molecular weight that are commercially available as a residue of petroleum or coal-tar refining. Its two major components are asphaltene compounds, which give bitumen colloidal properties, and malthene compounds, which impart viscous liquid properties. Most bitumen is obtained during the distillation of crude oil and is called "direct distilled" bitumen. This bitumen has a high viscosity and must be heated to make it suitable for mixing. The bitumen used in the CRL thin-film evaporator has been emulsified in water (contains 45% H_2O) and can be used without being heated.

OPERATIONS PERFORMANCE OF TFE-1

Effect of Rotor Change

Data have been collected from TFE-1 after a new rotor was installed and operated at higher speeds. Table 2 summarizes the performance of TFE-1 before and after the rotor change.

Table 2
Effect of Rotor Change On TFE-1 Performance

PARAMETER	PRE-ROTOR CHANGE	POST-ROTOR CHANGE	SUMMATION ALL DATA
VOLUME PROCESSED (L)	13160	13740	26900
NUMBER PRODUCT DRUMS	24.6	21.4	46
VOLUME REDUCTION FACTOR	2.67	3.21	2.92
AVERAGE VOLUME PER DRUM	535	642	585

The new rotor operates at a speed of 900 rpm, compared to 600 rpm prior to its installation. A total volume of 26.9 m^3 was immobilized with TFE-1 in 1994 with a volume reduction factor (VRF) of 2.92. A volume reduction factor (VRF) is defined by equation (1).

$$VRF_{TFE\text{-}1} = \frac{Feed\ Volume\ [TFE\text{-}1]}{Product\ Drum\ Volume} \qquad (1)$$

Prior to the installation of the new rotor a total volume of 13.2 m^3 of liquid had been processed with a VRF of 2.67. After the installation of the new rotor a total volume of 13.7 m^3 was processed with a VRF of 3.21. The cleaning frequency was reduced by a factor of three after the installation of the rotor operating at higher rpm. The higher VRF after the rotor change boosted the average volume of waste concentrate immobilized per drum from 535 L to 642 L. Over the duration of the year (including data for both rotors) an average volume of 585 L of waste concentrate was immobilized per drum. This represents an overall VRF of 2.92. On average over the duration of the year, there was about 64.2 kg of waste solids immobilized in each drum. The drums contained approximately 30.3% solids by weight. Inert solids (associated with grit and suspended matter removed by microfiltration in the backwash stream) represent about 4 to 5% of the weight of the product drum. The immobilized waste solids are comprised primarily of sodium nitrate, sodium phosphate, sodium sulphate, and sodium carbonate. The average concentrations of the primary radioactive and chemical contaminants immobilized in the product drums are given in Table 3. The data are expressed as a quantity of contaminant per unit mass of immobilized secondary waste, from TFE-1.

Table 3

Chemical and Radiological Characteristics of Drums

	PARAMETER	AVERAGE
RADIOLOGICAL	GROSS BETA	2.57E+00
CONTAMINANTS	AM-241	7.19E-03
	CE-144	9.14E-01
	CO-60	4.42E-01
(mCi/L)	CS-134	9.35E-02
	CS-137	9.01E-01
	EU-154	2.63E-02
	EU-155	1.57E-02
	NB-94	1.11E-03
	NB-95	2.25E-02
	RU-103	2.51E-03
	RU-106	9.78E-02
	SB-125	4.95E-03
	ZN-65	4.01E-02
	ZR-95	9.58E-03
	NI-63	2.46E-03
	PU-239/240	3.34E-03
	ALPHA	2.28E-02
CHEMICAL	PHOSPHATE	3.65E+04
CONTAMINANTS	CARBONATE	1.01E+04
	NITRATE	9.24E+04
(mg/L)	CHLORIDE	1.12E+04
	SULPHATE	1.84E+04
	SODIUM	6.10E+04
	IRON	1.77E+03
	CALCIUM	5.50E+02

Chemical characterizations of the ammonium hydroxide and citric acid-based cleaning solutions used for TFE-1 (prior to the installation of the new rotor), have shown that there is severe scaling of the evaporator by phosphate- and sulphate-based salts. About 6 kg of sodium phosphate and 1 kg of sodium sulphate were deposited on the 1 m^2 of evaporator surface area. Significant quantities of calcium, magnesium, iron, and silica were also detected in the ammonium citrate-based cleaning solution. Visual observations indicated that large deposits of hardened scale were present on the heat transfer surfaces, particularly within about 25 cm of the feed inlet. These observations indicated that all of the water associated with the waste solids was flashed off within approximately 0.3 m of the TFE-1 feed inlet. The length of the heated surface is 1.5 m.

Crusty deposits of scalable species are thought to be responsible for the reduction of the heat transfer area, and lower product temperatures prior to the installation of the new rotor. Product temperatures were recorded at one hour intervals during a processing campaign, which typically lasted approximately 14 hours. At the start of a campaign a process temperature of 155 °C was attainable but could not be maintained at this level for the duration of the run, due to the chemical scaling.

The chronic fouling observed prior to the rotor change has now been minimized. The frequency of chemical cleaning has been reduced by a factor of three after installation of the new rotor and this has minimized down time. It is now possible to maintain an outlet product temperature of between 135 - 145 °C for the duration of a 4000 L campaign. This ensures that there is no residual liquid in the product drum. There is less than 1% free water in a product drum based on mass balance.

For operations with the old rotor, the average combined distillate rate (evaporated from the waste and the bitumen) varied between 0.4 and 1.0 L/min. When the new rotor was initially installed the distillate rate increased to about 1.5 L/min, before subsequently decreasing back to an average of 1.0 L/min. However, the present distillate rate of 1.0 L/min can be maintained at an average product temperature of about 135°C over the duration of a 14 hour campaign.

Decontamination Factor of Radionuclides Treated by TFE-1

The removal efficiency of a radionuclide or other chemical contaminant in an evaporator is usually expressed as a decontamination factor (DF), which is defined by equation (2).

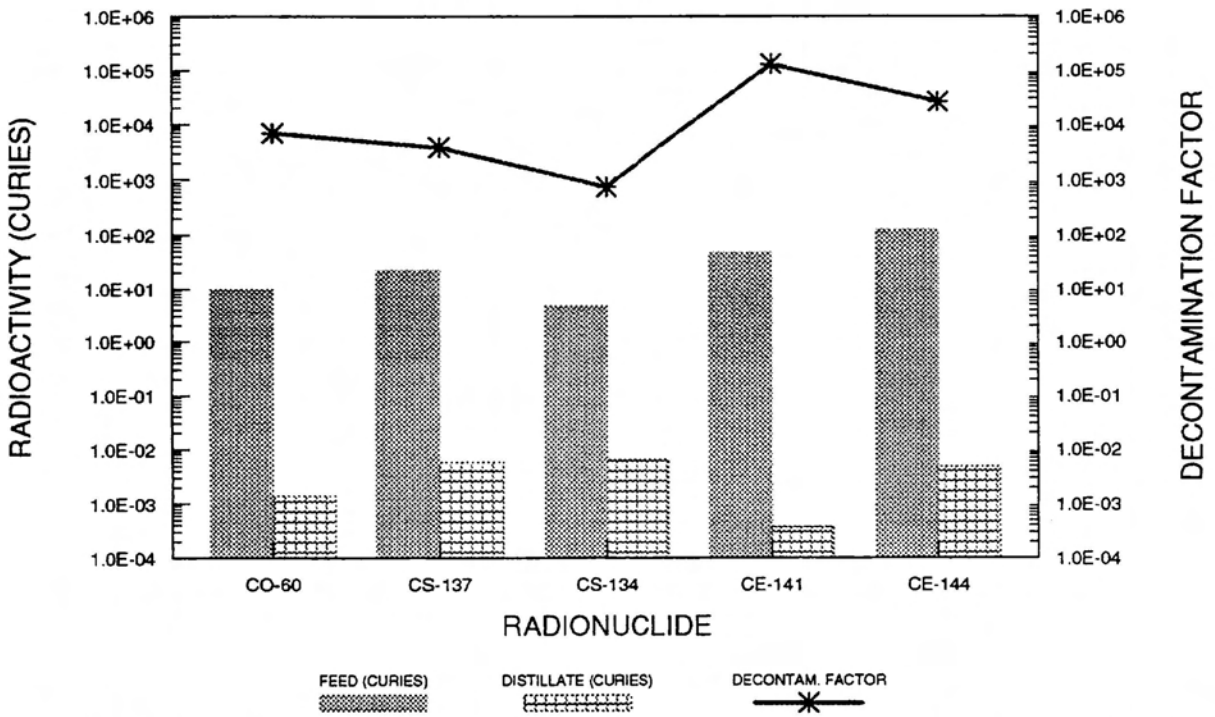
$$DF = \frac{[\beta/\gamma]_{feed}}{[\beta/\gamma]_{effluent}} \qquad (2)$$

Figure 2 shows the evaporator performance for the removal of the most abundant isotopes present in the feed stream to the evaporator. The data were compiled from over 200 campaigns. The first bar of each histogram represents the total radioactivity of the specified radionuclide in the feed stream in curies. The second bar is the total radioactivity of the radionuclide in the distillate stream, also in curies. Finally the line plot, which refers to the right ordinate, is the decontamination factor for each radionuclide.

Fig. 2

Decontamination Factors for Radionuclides

Radioactivity Treated with TFE-1



The first set of two bars shows the evaporator performance for the removal of ^{60}Co. Over an operating period of about 500 days, about 10 Ci of ^{60}Co was introduced into the solidification plant. Of this, less than 0.002 Ci was released, representing a DF of about 6000 in the thin-film evaporator. If the fraction of suspended particles to the evaporator for a given batch of waste is high it is probable that a large removal of ^{60}Co will be achieved, since a large fraction of this radionuclide is adsorbed to the surfaces of suspended particles.

The most problematic radionuclides from a decontamination perspective are ^{137}Cs and ^{134}Cs. About 80% of the cesium is present as ^{137}Cs, but both isotopes are equally difficult to remove upstream of the evaporators in the membrane plant, due to their low valence. The cesium isotopes are rejected at about 98% by the membrane plant, compared with 99.5% for the other β/γ emitters [8]. The bulk of the cesium is soluble and is not retained by the suspended solids in the backwash concentrate stream. The decontamination factors for ^{137}Cs and ^{134}Cs are 4000 and 800 respectively.

The decontamination factors for the two cerium isotopes (^{141}Ce and ^{144}Ce) are 10^5 and 2×10^4 respectively. The large DF for these two isotopes is partially attributed to the fact that the bulk of the cerium is adsorbed on suspended solids removed by the MF system. Since the retention of suspended solids in the evaporators is very high, it follows that there is very efficient removal of the cerium isotopes.

The lower DF for the two cesium isotopes may be related to the fraction of total cesium that is dissolved. By comparison with ^{141}Ce and ^{144}Ce, where the decontamination factors are greater than 20 000, and ^{60}Co (DF $\approx$ 6000), most of the cesium is dissolved. About 60% of the cobalt isotopes and 75% of the cerium isotopes are associated with the suspended solid phase. These adsorbed

radionuclides are subsequently removed by the MF membranes as backwash concentrate. The results suggest that the removal efficiency of the evaporators for radioactivity decreases when the dissolved fraction of the isotope increases. It is known from operating experience at CRL that, of the radionuclides discussed here, the cerium isotopes are the least soluble, and the cesium isotopes are the most soluble.

ORGANIC CARRYOVER IN EVAPORATOR DISTILLATE

Although the effluent quality from the evaporator meets all discharge criteria for radioactivity, there is some carryover of organics in the distillate. The source of these organics is primarily the distillation of lighter components in the emulsified bitumen in TFE-1, which is used for immobilization of the mixed aqueous waste concentrate. The concentration of dissolved organic carbon (DOC) in the distillate can vary between 1 and 300 mg/L, while the concentration of phenolics in the distillate stream ranges from 0.02 to 2 mg/L. The oil and grease concentration in the distillate stream is usually less than the 15 mg/L Federal Discharge target [3], but has exceeded 100 mg/L on occasion.

It has been observed that a titanium dioxide photocatalytic reactor is capable of reducing both EPA 624 (volatile) and EPA 625 (extractable) priority contaminants present in the evaporator distillate to below the method detection limits of the GC/MS analytical equipment [4]. Dissolved oxygen was sufficient to remove the colour associated with organics from the wastewater, even though not all of the organic carbon was removed. A concentration of 500 mg/L of hydrogen peroxide was found to be sufficient for the removal of the organics present in the evaporator distillate stream. Phenolics were effectively reduced to well below the Canadian Federal Discharge limit of 20 µg/L with the technology, and oil and grease was reduced to below the 15 mg/L imposed guideline. Other aromatic compounds, including naphthalene and methyl-substituted naphthalene derivatives, were removed or converted to other less toxic constituents. The dissolved organic carbon was not all converted to carbon dioxide in the catalytic reactor; some intermediate oxidation products were formed, which included organic acids.

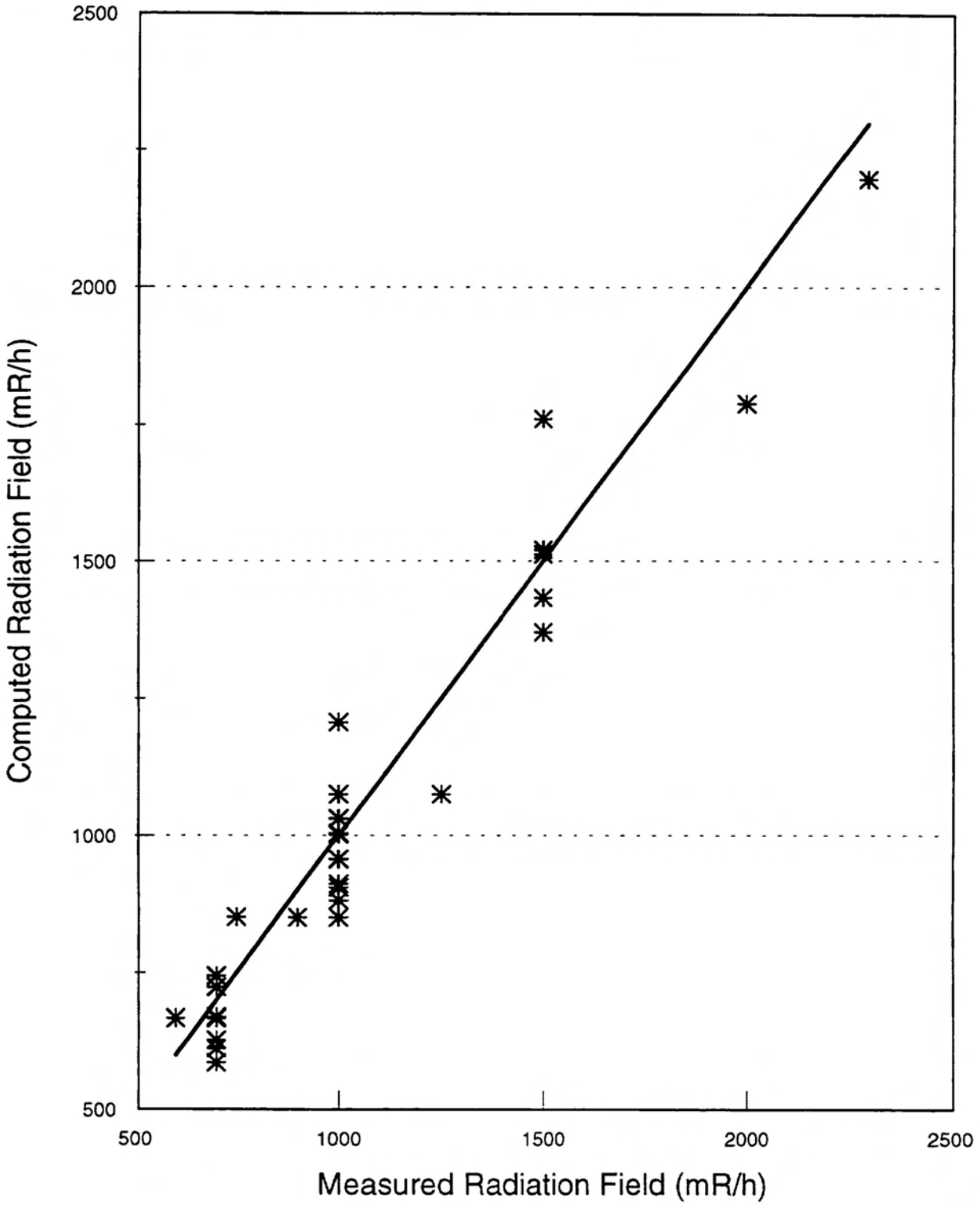
PRODUCT DRUM RADIATION FIELD

The radiation field on the product drums is a function of the concentrations and distributions of the specific β/γ emitters immobilized in it. The contact field on a product drum varied between 100 and 1000 mR/h for the first 100 drums produced at the WTC, during which time the weight per cent solids in the product drum was maintained below 25%. Thereafter, with more radioactive waste introduced into the WTC, and by maintaining the weight per cent solids at about 35%, the average contact radiation field increased significantly. There have been two runs to date where 5 R/h (contact) drums were produced. The new rotor operating at the higher speed of 900 rpm has increased the volume reduction potential of the evaporator as discussed earlier (Table 2). This is because there is better heat transfer through the vessel wall.

A semi-empirical correlation was employed which allows the radiation field to be computed if the concentration of gamma emitters in the feed tank is known. Figure 3 shows the correlation between the radiation field (as measured on contact on the side of the drum), and the estimated radiation field. The correlation is accurate to within 7%, if the radiation field on contact with

Fig. 3

Estimation of Radiation Field

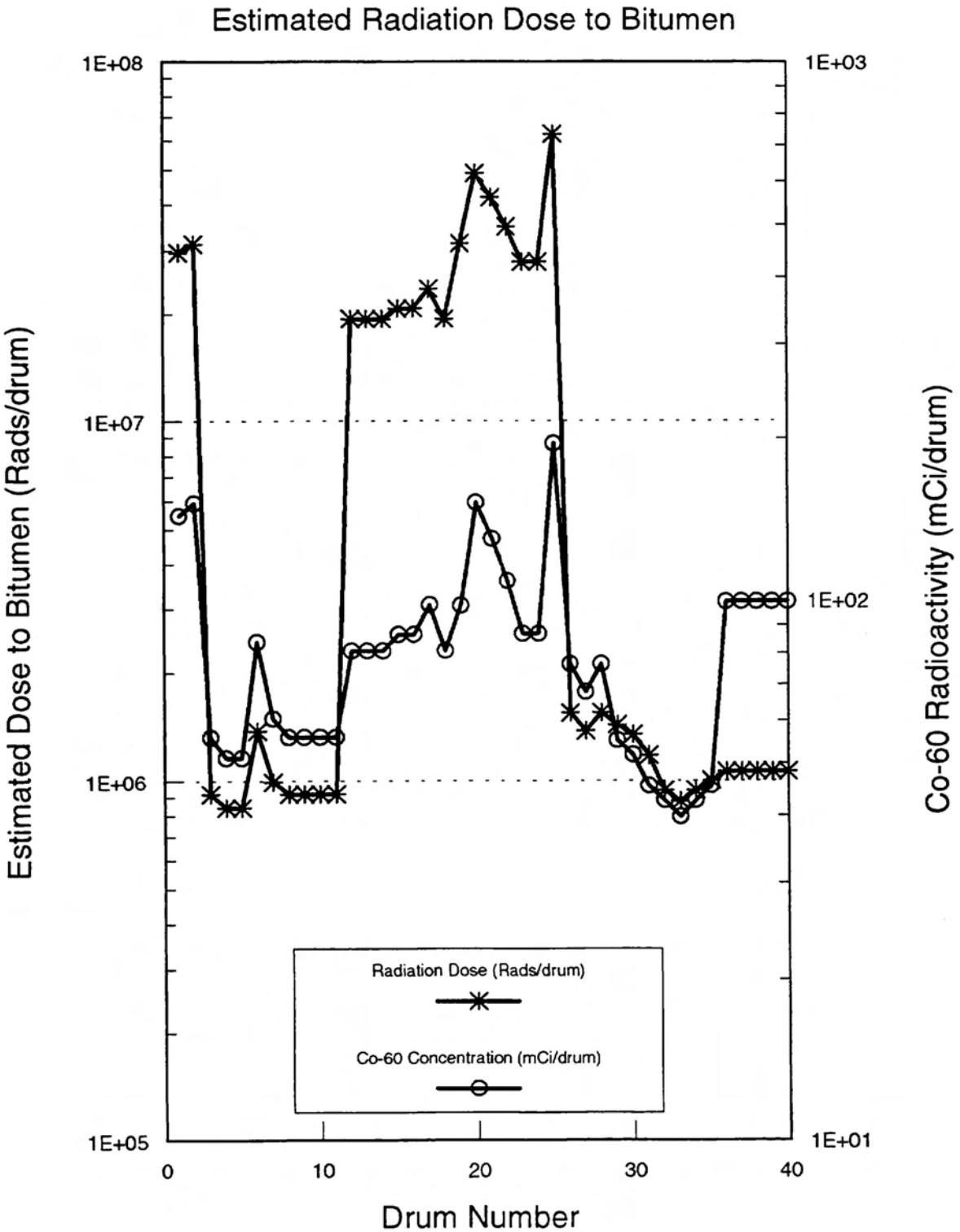


the drum exceeds 500 mR/h. The correlation is accurate enough to allow fine tuning of the liquid and bitumen emulsion flowrates. This ensures that the radiation field on any drum does not exceed about 3 R/h on contact.

The estimated radiation dose to the bitumen was computed over its lifetime. The gross beta/gamma loading per kg of product varied between 0.001 and 0.01 Ci/kg. This resulted in an estimated dose to the bitumen ranging between 1 x 10^6 to 1 x 10^8 rads per drum over its lifetime. This was well within the maximum allowable dose of about 5 x 10^8 rads per drum [7].

The estimated dose to the bitumen (from the known beta/gamma emitters) for the 40 product drums generated in 1994, is shown in Fig. 4 on the left ordinate. A large fraction of the dose to the bitumen is due to Co-60 radioactivity (shown on the right ordinate in mCi/drum).

Fig. 4


Estimated Radiation Dose to Bitumen
1E+08
1E+07
1E+06
1E+05
1E+03
1E+02
1E+01
0
10
20
30
40
Drum Number
Estimated Dose to Bitumen (Rads/drum)
Co-60 Radioactivity (mCi/drum)
Radiation Dose (Rads/drum)
Co-60 Concentration (mCi/drum)

RADIOLOGICAL AND CHEMICAL CHARACTERISTICS OF PRODUCT DRUM

The total inventory of radioactivity and salt loading immobilized in the 40 product drums was computed. These totals were divided by the total mass of immobilized secondary bitumen waste produced from the WTC in 1994. Table 3 shows the distribution of radiological and chemical contaminants expressed in the units of μCi/mL (of immobilized product) and mg/kg (of immobilized product), respectively. Of the β/γ emitters immobilized in the product drums, the most abundant were: Ce-144 (0.91 μCi/mL), Co-60 (0.44 μCi/mL), Cs-137 (0.90 μCi/mL), and Cs-134 (0.94 μCi/mL). On average, there was a total of 0.023 μCi/mL of gross α present, about half of which could be accounted for by the two radionuclides: Am-241 (0.0072 μCi/mL) and Pu-239 (0.0033 μCi/mL). The gross α accounts for about 1% of the total radioactivity in the product drum.

Five chemicals have been identified as contributing towards the majority of the reactive solids loading in the product drum. These include the sodium-based salts of nitrate (92 g/kg), phosphate (36.5 g/kg), sulphate (18.4 g/kg), chloride (11.2g/kg), and carbonate (10.1 g/kg). Concentrations of sodium, iron, and calcium are also given in Table 3.

CONCLUSIONS

Product drums from the thin-film evaporator facility at the CRL Waste Treatment Centre have an on-contact radiation field of between 500 mR/h and 3000 mR/h, depending upon the radiological and chemical characteristics of the feed. The dominant β/γ emitters in the product drum include: ^{137}Cs, ^{134}Cs, ^{144}Ce, ^{141}Ce, and ^{60}Co. On average, there is a gross β/γ loading of 2.6 μCi/mL in the immobilized bitumen product. The concentration of α emitters constitutes about 1% of the total radioactivity, and the principal isotope is ^{241}Am. The radiation dose to the bitumen varies between 10^6 and 10^8 rads per product drum (over its lifetime), which is less than 20% of the maximum allowable dose for the matrix.

The decontamination factor for various radionuclides in the thin-film evaporator varies from about 20 000 for ^{144}Ce to 6000 for ^{60}Co, and about 4000 for ^{137}Cs. Those radionuclides more strongly adsorbed to the inert solids associated with the MF backwash solids are more efficiently removed in the evaporator.

Installation of a new rotor, and increasing its speed from 600 rpm to 900 rpm, has significantly improved the operation of the thin-film evaporator used for immobilization. The cleaning frequency of the unit has been reduced by a factor of three, and the solids loading has been boosted from about 25% to 35%. The volume reduction factor increased from 2.67 to 3.21 after the installation of the new rotor operating at 900 rpm.

There is some distillation of the organic fraction from the emulsified bitumen used for immobilization of the mixed waste concentrate. Photocatalytic oxidation can effectively remove the organics volatilized in the distillate.

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