#### TESTING OF HIGHLY SELECTIVE CoTreat ION EXCHANGE MEDIA FOR THE REMOVAL OF RADIOCOBALT AND OTHER ACTIVATED CORROSION PRODUCT NUCLIDES FROM NPP WASTE WATERS

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### ABSTRACT

In the R&D of highly selective inorganic ion exchange media the major attention has been in the separation of <sup>137</sup>Cs, <sup>90</sup>Sr or Transuranium (TRU) nuclides and a good number of effective media are commercially available for the removal of these radionuclides from nuclear waste effluents. Less attention has been paid to the selective separation of other, shorter-lived radionuclides. CoTreat is a new inorganic media (manufactured by Selion OY, Finland) that is highly selective for <sup>58,60</sup>Co and other activated corrosion product nuclides (<sup>54</sup>Mn, <sup>59</sup>Fe, <sup>65</sup>Zn, <sup>63</sup>Ni).

An extensive test programme has been carried out at laboratory scale to evaluate CoTreat performance for the purification of real Floor Drain and Equipment Drain waters from several nuclear power plants (NPP) in Finland (Loviisa NPP, Olkiluoto NPP) and in the US (Diablo Canyon NPP, Ginna NPP, Harris NPP). The tests included filtration tests (filter pore size 5.0-0.005  $\mu$ m), determination of batch distribution coefficient (k<sub>d</sub>) for the radionuclides present in the water samples and small-scale column tests (bed volume 0.5 ml). In cases that the water samples contained also <sup>137</sup>Cs (Loviisa, Ginna, Harris), CsTreat<sup>®</sup> column was used in series with the CoTreat column. For the Loviisa sample, two other commercial media, active carbon (Norit RS) and an organic resin (Diphonix) were tested in parallel as a reference.

The filterability of the radionuclides was very different in the water samples. For instance, in the Loviisa sample, the filtration at 0.45 µm gave a high decontamination factor (DF) for most radionuclides except <sup>137</sup>Cs (<sup>54</sup>Mn DF = 40, <sup>60</sup>Co DF = 28, <sup>58</sup>Co DF = 11, <sup>110m</sup>Ag DF = 27) so these radionuclides were present mainly in a solid form. In other samples (Ginna 1, Harris), a major proportion of radionuclides were present in soluble form and low DF's (DF<2) were obtained even with UF (0.005  $\mu$ m). The measured k<sub>d</sub>-values, which are theoretically identical with the maximum column capacities (l/kg) for the radionuclides increased as pH was increased and were high, in the range of  $50,000 - 500,000 \text{ L/kg} (300,000 - 3,000,000 \text{ gal/ft}^3)$  at neutral pHvalues (pH 6-8) for <sup>58,60</sup>Co, <sup>54</sup>Mn and <sup>65</sup>Zn. High DF's (300-500) were obtained in the column test for the radionuclides in samples with soluble radionuclides and the DF's were lower (10-50) in cases that most of radioactivity was initially present in solid form. However, even in these cases, the over-all DF's (filtration + CoTreat column) were generally high (DF 100-500). These labscale tests indicate that CoTreat media is highly efficient for the removal of radiocobalt and other activated corrosion nuclides from NPP wastewaters, outperforming several other commercial Coselective media. Larger scale tests of CoTreat (125 ml column) are underway at several US NPP sites.

# **INTRODUCTION**

Due to the increasing costs of final disposal, nuclear power plants are taking action to decrease their radwaste volumes. The experience gained in several utility sites is showing that replacing evaporators or conventional demineralizer resins in liquid radwaste treatment systems by highly selective ion exchange media can bring considerable reductions in radwaste volumes and treatments costs [1-4]. At present, however, commercial highly selective media (e.g. zeolites) are available mainly for the removal of <sup>137</sup>Cs, which is often the most critical nuclide in the liquid radwaste. However in many plants <sup>60</sup>Co and <sup>58</sup>Co are more critical than <sup>137</sup>Cs and also the removal of other shorter-lived radionuclides like <sup>54</sup>Mn, <sup>59</sup>Fe, <sup>63</sup>Ni, <sup>65</sup>Zn, <sup>110m</sup>Ag and <sup>124,125</sup>Sb is becoming important.

Worldwide, in the research of selective removal of radionuclides from nuclear waste solutions the main emphasis has been on two radionuclides, namely <sup>137</sup>Cs and <sup>90</sup>Sr. Historically, this is a reflection of the extensive pioneering work that was carried out in the US National Laboratories (Oak Ridge, Savannah River, Los Alamos) for the treatment of medium- or high-active waste solutions, in which <sup>137</sup>Cs, <sup>90</sup>Sr and TRU nuclides are abundant. As the results of the R&D work, several efficient materials are available commercially today for the separation of these radionuclides. Zeolites were the first materials to be used in large-scale processes in NPP's and nuclear fuel reprocessing plants. However, the selectivity of zeolites is not high enough to give a satisfactory processing capacity for medium- and high-salt solutions. Therefore, more selective media (e.g. titanates, silicotitanates and hexacyanoferrates) have been developed for separation of <sup>137</sup>Cs and <sup>90</sup>Sr. Most selective of these is the CsTreat<sup>®</sup> media, which is presently used in several NPP's and other nuclear sites worldwide [2].

Relatively little R&D has been carried out for the selective removal of other radionuclides, of which especially activated corrosion products (e.g. <sup>54</sup>Mn, <sup>58,60</sup>Co, <sup>59</sup>Fe, <sup>63</sup>Ni, <sup>65</sup>Zn) are important. Many chelating organic resins, developed originally for non-radioactive applications, are likely to be selective in some extent for certain nuclides. Some new resins have been also developed with nuclear applications in mind, e.g. resins with phosphonic acid functionality. Also many active carbons are considered selective for <sup>60</sup>Co and other trace metal ions. Highly selective inorganic media, having the advantage of higher radiation and thermal stability compared to organic resins have been in general absent in the arsenal for the removal of radiocobalt.

#### **EXPERIMENTAL**

For the tests, samples from the Floor and Equipment Drain Collection Tanks were obtained from four utility sites: Loviisa (Finland), Ginna (USA), Diablo Canyon (USA) and Harris (USA). Loviisa supplied three 5 gal. samples, Ginna supplied one 5 gal. sample and Diablo Canyon two 1 gal. samples. A two gallon sample was obtained from Harris. The sites carried out the initial chemical (Loviisa) and/or radiochemical analyses (all sites) of the samples (see Tables I & II).

Component	Concentration
Na	36.8 mg/l
Κ	7.7
Са	0.86
H <sub>3</sub> BO <sub>3</sub>	160
pH	8.7-9.0
Mn-54	111 Bq/l
Co-58	27
Co-60	402
Ag-110m	203
Sb-124	33
Cs-134	274
Cs-137	432

Table I. Chemical and radiochemical composition of Floor Drain Water, Loviisa NPP, Finland

Nuclide				
	Ginna	Diablo Canyon 1	Diablo Canyon 2	Harris
Cr-51	8,550			
Mn-54	1,830	2,700	69,000	3,380
Co-58	107,000	422,500	49,500	1,700
Co-60	15,200	6,500	443,000	8,300
Sb-125	2,300	1,600	44,000	860
Ag-110m	9,300			
Cs-134	1,300			1,500
Cs-137	6,050			5,700
Ni-63			185,000	

Three selective media, CoTreat (manufactured by Selion OY, Finland), active carbon Norit RS (Norit, Netherlands) and an organic resin Diphonix (Eichrom Industries, USA) were tested in parallel for the corrosion product radionuclide separations. Because the liquid samples contained <sup>137,134</sup>Cs and CoTreat does not take up cesium appreciably, CsTreat<sup>®</sup> ion exchange media tested also for cesium removal. CoTreat is a new all-inorganic, titanium-based ion exchange media. Diphonix resins has dual sulphonic acid/aminophosphonic acid functionality. CsTreat is a hexacyanoferrate-based all inorganic media. All media were used in granular form. The tests comprised of filtration tests, static batch experiments and column experiments. Due to the limited sample volume obtained from the US sites, Norit RS and Diphonix were tested only for the Loviisa samples.

All water samples had visibly solid material in the bottom of the sample bottles and some also suspended solid material. In the filtration tests, the water samples were filtered using 5.0  $\mu$ m, 0.45  $\mu$ m and 10,000 Dalton (~0.005  $\mu$ m) membrane filters in parallel, i.e. each solution sample was filtered only once.

In the batch experiments the distribution coefficient ( $k_d$ ) of different radionuclicdes was determined at the original pH of the samples (5.6-6.2) and at slightly adjusted pH  $\approx$  7, which had been found an average optimum for the different radionuclides in phase one tests. Water samples were filtered with a 0.45 µm membrane prior to the determinations. The  $k_d$ -determinations were

carried out by equilibrating the media with the solution samples (100 mL of solution/0.01 g of media) in rotated plastic bottles for two weeks. After equilibration, the solution was centrifuged, filtered and the radionuclide concentrations were analyzed. The values of  $k_d$  were calculated using a formula:

$$\mathbf{k}_{d} = \frac{\overline{\mathbf{A}}}{\overline{\mathbf{A}}} = \frac{\left(\mathbf{A}_{0} - \mathbf{A}\right)}{\overline{\mathbf{A}}} \frac{\mathbf{V}}{\mathbf{m}}$$
(1)

where  $\overline{A}$  and A are the equilibrium activity concentrations of the radionuclide in the exchanger and in the solution, respectively, A<sub>0</sub> is the activity concentration initially, V is the solution volume (100 mL) and m is the exchanger mass (0.01 g). The high V/m ratio of 10,000 mL/g was used to avoid changes in the solution composition of the inactive ions. The k<sub>d</sub>- results are thus representing the same equilbria that would be attained in column operation. Under these circumstances, the k<sub>d</sub>-value also gives the maximum theoretical column capacity, in terms of solution volume (L) that can be purified per unit mass (kg) of exchanger<sup>1</sup>.

Column experiments were carried out using small lab-scale columns (bed volume 0.5 mL.). Water samples were filtered (a 0.45  $\mu$ m) before the column runs, and the water pH was adjusted to neutral in cases that the original pH was slightly acidic or alkaline. The flow rate was 20-25 bed volumes/hour and exchanger grain size 0.85-0.25 mm. Column effluent was collected as fractions, which were analysed for the activity concentration of different radionuclides.

# RESULTS

## **Filtration Tests**

In Loviisa Floor Drain (FD) water (sample No.1), large proportion all radionuclides except  $^{134,137}$ Cs could be removed by filtration (Table III). The lowest DF's were obtained for  $^{58}$ Co (DF = 8.3 - 12.8) and the highest for  $^{54}$ Mn (DF = 37-42). The DF's increased only slightly as the pore size was decreased from 5.0 µm to 10,000 D (~ 5.0 nm), i.e. the pore size was decreased 1000-fold. This shows that large majority of  $^{54}$ Mn,  $^{58,60}$ Co, and  $^{110m}$ Ag were bound to particles larger that 5.0 µm. The DF's were clearly higher for  $^{60}$ Co than for  $^{58}$ Co, i.e. larger proportion of  $^{60}$ Co was in particle form. This propably reflects the different origin of these nuclides.

In Ginna FD water, all <sup>51</sup>Cr was removed by filtration with 0.45  $\mu$ m and 10,000 D membranes. <sup>54</sup>Mn and <sup>60</sup>Co were removed with a DF greater than 10 and the DF was 2-5 for the rest of the radionuclides excluding <sup>137</sup>Cs (DF  $\approx$  1) and <sup>110m</sup>Ag. A very high DF of 260 was obtained for <sup>110m</sup>Ag with the ultrafiltration membrane.

In the Diablo Canyon sample No. 1 and in the Harris sample, the DF's obtained by filtration were lower than 2 indicating that the radionuclides were mainly in a soluble form. Diablo Canyon sample No. 2 had similar characteristics than the Ginna sample, filtration removed <sup>54</sup>Mn and <sup>60</sup>Co with a relative good DF (8-32), while <sup>57,58</sup>Co and <sup>125</sup>Sb were removed with a modest DF.

# **Distribution Coefficients**

In Loviisa FD water, distribution coefficient  $k_d$  of the media could be determined for <sup>60</sup>Co and <sup>137</sup>Cs only (also for <sup>54</sup>Mn in case of Norit RS), since after filtration, the activity concentration of Table III. Decontamination factor for various radionuclides obtained in filtration tests

Sample	DF		
Nuclide	Filter pore size		
LOVIISA SAMPLE 1	5.0 µm	0.45 μm	10,000 D
Mn-54	37.	4 39.8	41.5
Co-58	8.	3 10.8	12.8
Co-60	31.	4 27.7	29.3
Ag-110m	16.	2 27.0	BDL
Cs-137	1.	0 1.0	1.2
GINNA	5.0 µm	0.45 μm	10,000 D
Cr-51	NA	BDL	BDL
Co-57	NA	4.5	5.3
Mn-54	NA	25.9	40.4
Co-58	NA	3.1	3.2
Co-60	NA	12.1	15.7
Zn-65	NA	3.4	4.3
Ag-110m	NA	3.1	258.7
Sb-125	NA	2.2	2.2
Cs-134	NA	1.0	1.1
DIABLO CANYON 1	5.0 µm	0.45 μm	10,000 D
Mn-54	1.	5 1.7	1.4
Co-58	1.	6 1.6	1.0
Co-60	1.	6 2.0	1.8
Sb-125	1.	0 1.0	1.0
DIABLO CANYON 2	5.0 µm	0.45 μm	10,000 D
Mn-54	4.	2 9.9	23.5
Co-58	4.	0 5.0	7.8
Co-60	4.	0 8.0	32.0
Ag-110m	BDL	3.4	BDL
Sb-125	2.	8 3.4	3.6
HARRIS	5.0 µm	0.45 μm	10,000 D
Mn-54		1 1.1	1.1
Co-58	1.	1 1.4	1.2
Co-60	1.	4 1.8	1.7
Sb-125		1 1	1
Cs-137		1 1.1	1.3

BDL = not detected, NA = not analyzed

other radionuclides was very low and fell below detection limit in the batch contact with the media. For CoTreat, the  $k_d$  of  ${}^{60}$ Co had a maximum of 59,000 L/kg (350,000 gal/ft<sup>3</sup>) in neutral pH (6.71)(Table IV.). In slightly alkaline solution the  $k_d$  was considerably lower, which is propably

due to the formation of hydrolysis products like  $Co(OH)^+$  and  $Co(OH)_2$  (aq), which are likely to decrease the sorption of divalent cobalt ions to the exchanger. In moderately acidic solution (pH = 3.21) the k<sub>d</sub> was low, reflecting the weak-acid nature of the CoTreat material. Surprisingly, CoTreat material also showed rather good k<sub>d</sub>-values for <sup>137</sup>Cs.

Norit RS showed similar  $k_d$ -values to CoTreat for <sup>60</sup>Co, except that in neutral pH, the value was clearly lower, while Diphonix resin had considerably higher  $k_d$ 's than CoTreat. Based on this  $k_d$ -data it would have been expected that Diphonix would outperform CoTreat in the column removal of <sup>60</sup>Co. However, this was not the case (see later).

In Ginna FD water, the  $k_d$ 's (CoTreat tested only) were considerably higher in neutral pH (6.95) than is slightly acidic pH (5.60) for all nuclides except <sup>125</sup>Sb (TableV). In general, the  $k_d$ 's for <sup>60</sup>Co were higher than for <sup>57</sup>Co and <sup>58</sup>Co, while the <sup>57</sup>Co and <sup>58</sup>Co  $k_d$ 's were close to equal. This

Sample/Media			K <sub>d</sub> (L/kg)	
Nuclide			рН <sub>еq</sub>	
LOVIISA SAMPLE 1/CoTreat	3.21		6.71	8.29
Cs-137		10,500	28,000	16,600
Co-58		BDL	BDL	BDL
Mn-54		BDL	BDL	BDL
Ag-110m		BDL	BDL	BDL
Co-60		3,600	59,000	20,200
LOVIISA SAMPLE 1/Norit RS	3.09		6.41	8.16
Cs-134		4,700	4,300	0
Co-58		BDL	BDL	BDL
Mn-54		1,900	9,800	0
Ag-110m		BDL	BDL	BDL
Co-60		3,800	22,000	22,000
LOVIISA SAMPLE 1/Diphonix	3.01		4.40	7.91
Cs-134		18,400	1,300	3,700
Co-58		BDL	BDL	BDL
Mn-54		BDL	BDL	BDL
Ag-110m		BDL	BDL	BDL
Co-60		130,000	187,000	82,000

Table IV. Distribution coefficients  $k_d$  for various radionuclides and media obtained in batch equilibration tests with Loviisa sample 1.

MDA = activity below detection limit

trend is opposite of what one would expect based on the filtration results, which indicated that  ${}^{60}$ Co was present in the particle (non-ionic) form to a larger degree than  ${}^{57}$ Co and  ${}^{58}$ Co. In neutral pH, the k<sub>d</sub>-values are very high for  ${}^{110m}$ Ag (190,000 L/kg) and  ${}^{65}$ Zn (158,000 L/kg) and high for the different nuclides of cobalt (31,000 – 49,000 L/kg) and  ${}^{54}$ Mn (57,000 L/kg).

For Diablo Canyon Equipment Drain Waters, very high  $k_d$ 's were obtained (Table V) for radiocobalt nuclides (765,000-65,000 L/kg) and for <sup>54</sup>Mn (300,000 L/kg). These  $k_d$ -values were also very high for the harris sample. Diablo Canyon sample No. 2 contained a high level of <sup>125</sup>Sb and a rather low  $k_d$ , similar to the Ginna water, was obtained for that nuclide (5,500 L/kg).

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Sample	K <sub>d</sub> (L/kg)		
Nuclide	pF	pH <sub>eq</sub>	
GINNA	5.60	6.95	
Co-57	6,300	31,600	
Cr-51	BDL	BDL	
Sn-113	BDL	BDL	
Sb-125	11,570	5,000	
Zr-95	BDL	BDL	
Cs-134	3,900	4,600	
Co-58	8,700	36,600	
Mn-54	13,000	57,000	
Ag-110m	64,000	190,000	
Zn-65	26,300	158,000	
Co-60	11,500	49,000	
DIABLO CANYON 1	6.08		
Sb-125	BDL		
Co-58	765,000		
Mn-54	BDL		
Co-60	633,000		
DIABLO CANYON 2	6.34		
Co-57	BDL		
Sb-125	5,500		
Co-58	390,000		
Mn-54	307,000		
Ag-110m	26,000		
Co-60	78,000		
HARRIS	6.85		
Mn-54	600,000		
Co-58	335,000		
Co-60	215,000		
Sb-125	0		

Table V. Distribution coefficients  $k_d$  for various radionuclides and CoTreat obtained in batch equilibration tests with the Ginna and Diablo Canyon samples.

BDL = not detected

## **Column Tests**

In Loviisa FD water, the CoTreat column DF for <sup>60</sup>Co was almost constant at about 10 and the column exhausted at about 50,000 bed volumes (Fig. 1). The DF's for <sup>54</sup>Mn were initially high (<sup>54</sup>Mn below detection limit), but stabilized later to a level of 6-7. <sup>110m</sup>Ag DF's were somewhat lower (2-5). The CsTreat column DF's for <sup>137</sup>Cs were in the range of 335-711, and there was no sign of column exhaustion for <sup>137</sup>Cs at 50,000 BV.

In the Diphonix column, the DF's for <sup>60</sup>Co fell steadily from the initial value of 25 to 1.5 at 15,000 BV; i.e. the column was exhausted at 15,000 BV (Fig. 2). The performance of Norit RS column was very poor as the column was exhausted after about 2,000 BV. Based on the  $k_d$ -data in Table IV, it was surprising that the column performance of Diphonix was lower than that of CoTreat. Norit RS performed also much poorer than what the  $k_d$ -values indicated. CoTreat, on



Figure 1. Column DF for different radionuclides as a function of bed volumes processed (BV) Loviisa Floor Drain Water. Water was prefiltered (0.45  $\mu$ m) before column run. One CoTreat column and one CsTreat column in series (each BV = 0.5 ml). Flow rate 12-13 mL/h (24-26 BV/h).



Figure 2. Comparison of column DF's for <sup>60</sup>Co vs. bed volumes (BV) processed for different media in Loviisa Floor Drain Water.

the other hand, operated as expected from the  $k_d$ -values. The  $k_d$ -value of <sup>60</sup>Co in Table III suggested that the maximum column capacity of CoTreat would be 59,000 L/kg or 47,000 BV (CoTreat bulk density 0.8 kg/L) at pH = 6.71. The water pH was maintained at 6.5-7.0 in the column run, i.e. close to the pH at which the 47,000 BV capacity was measured batchwise and the observed column capacity of about 50,000 BV (Fig. 1) is in excellent agreement with this value. The differences in the column capacities and  $k_d$ -values for different media may be reflecting the different mechanisms of uptake. The assumption of equivalence of  $k_d$  and maximum column capacity can be derived for pure ion exchange reactions. CoTreat is a classical ion exchanger and is expected to obey this rule. Diphonix resin and the active carbon may have different sorption mechanisms, e.g. Diphonix contains chelating aminophosphonic groups and also active carbons are known to contain various organic groups of chelating function. The simple  $k_d$ /column capacity relationship may not be valid for uptake by chelate formation.

In Ginna FD water, the CoTreat column performance was lower (Fig. 3) than in the Loviisa case although the  $k_d$ -values (Tables III-IV) indicated column capacities of similar magnitude. Radionuclide levels in Ginna water were much higher than in Loviisa water, but in both cases the chemical concentrations of the radionuclides were so low that they should have no effect on the column capacity. Lower capacities for the Ginna sample obviously due to the different water pH. Although water pH was adjusted neutral initially it soon drifted to 5.7-5.9 due to the uptake of carbon dioxide from the air. In the Loviisa water there was no such strong decrease in pH from the adjusted value, propably due to the buffering effect of boric acid/borate, and the operating pH could be kept at a higher level of around 6.5. In Ginna water, the column was exhausted for <sup>58,60</sup>Co at about 10,000 BV (12,000 L/kg), which is practically identical with the  $k_d$ -value of 11,500 L/kg measured at pH = 5.60 (in the column run the pH was 5.7-5.9). If an effort had been made to maintain the operating pH at about 7, a much higher capacity of around 50,000 L/kg (40,000 BV) should have been obtainable. The column capacity for <sup>65</sup>Zn was high as expected from the  $k_d$ -value.

Diablo Canyon samples were only run to about 4000 BV due to the limited sample volumes. No sign of column exhaustion was observed at this volume for any of the radionuclides (Figs. 4 and 5). In sample No. 1, high DF's (300-600) were obtained for  ${}^{60}$ Co and  ${}^{58}$ Co.  ${}^{125}$ Sb was removed with a DF of 7-12. In sample No. 2 the CoTreat column was less efficient, the DF for  ${}^{58}$ Co was 40-50 but for  ${}^{60}$ Co only 4-6.  ${}^{54}$ Mn was removed with a DF of about 10. There was no uptake of  ${}^{125}$ Sb.

Harris sample was run until about 16,000 BV at which point all sample had been used (Fig. 6). The DF's for <sup>58,60</sup>Co and <sup>54</sup>Mn were above 100 initially but had fallen to about 10 at 16,000 BV. At this point, the column was still removing more than 90 % of these nuclides, i.e. the column still had considerable processing capacity left. The CoTreat column did not remove any <sup>125</sup>Sb. CsTreat column removed <sup>137</sup>Cs with a good DF (~ 1000) with no sign of column exhaustion at 16,000 BV.

It should be noted that the DF-values discussed above were obtained using very small lab-scale columns in which the column diameter/particle size ratio was very low. Under these circumstances channeling effects are likely to decrease the DF-values. It is expected that larger

columns will be able to produce higher DF-values.



Figure 3. Column DF for different radionuclides as a function of bed volumes processed (BV) Ginna Floor Drain Water. Water was prefiltered (0.45  $\mu$ m) before column run. One CoTreat column and one CsTreat column in series (each BV = 0.5 ml). Flow rate 12-13 mL/h (24-26 BV/h). The activity of <sup>137,134</sup>Cs was below the detection limit in most fractions collected. Successful measurements gave DF's in the range of 700-5000 for <sup>137,134</sup>Cs.



Figure 4. Column DF for different radionuclides as a function of bed volumes processed (BV) Diablo Canyon Floor Drain Water (sample No. 1). Water was prefiltered (0.45  $\mu$ m) before column run. One CoTreat column and one CsTreat column in series (each BV = 0.5 ml). Flow



rate 12-13 mL/h (24-26 BV/h).

Figure 5. Column DF for different radionuclides as a function of bed volumes processed (BV) Diablo Canyon Floor Drain Water (sample No. 2). Water was prefiltered (0.45  $\mu$ m) before column run. One CoTreat column and one CsTreat column in series (each BV = 0.5 ml). Flow rate 12-13 mL/h (24-26 BV/h).



Figure 6. Column DF for different radionuclides as a function of bed volumes processed (BV) for Harris NPP Floor Drain Water. Water was prefiltered (0.45  $\mu$ m) before column run. One CoTreat column and one CsTreat column in series (each BV = 0.5 ml). Flow rate 12-13 mL/h (24-26 BV/h).

## SUMMARY

The new CoTreat material appears very efficient for the removal of activated corrosion product nuclides from NPP wastewaters. The  $k_d$ -measurements indicate that processing capacities in the range of 300,000 – 3,000,000 gal/ft<sup>3</sup> should be obtainable for the removal of <sup>58,60</sup>Co. In order to obtain higher capacities it is necessary to control the water pH at neutral value. Column data obtained to date supports these capacity estimations for cobalt nuclides made by the  $k_d$ -measurements.

Because in most cases the corrosion product radionuclides were present mostly in particle form, the treatment process for the waste liquids must contain a prefiltration stage. Although in some cases the DF's obtained in column operation were not very large, the combined DF obtainable by filtration and column exchanges became high (Table VI). The DF's were very variable in different water samples, but even in the worst cases DF's in the range of 30-50 are obtained for <sup>60</sup>Co and in best cases the DF is in the order of 1000. <sup>54</sup>Mn and <sup>65</sup>Zn were also removed with a good DF. The DF's for <sup>110m</sup>Ag and <sup>125</sup>Sb were rather modest. Very high DF's (335-5000) were obtained for <sup>137</sup>Cs with the CsTreat media.

Based on the capacities and DF-values measured in these tests, the use of filtration and CoTreat/CsTreat media appears to be a highly efficient method for the treatment of wide range of NPP waste waters.

NUCLIDE/SAMPLE	OVERALL DF
LOVUSA No. 1	36,000 BV processed test continues
Mn 54	240 270
Co. 59	240-270 DDI
C0-58	
	280
Ag-110m	54-135
Cs-137	335-711
GINNA	Test terminated at 20,000 BV
Cr-51	BDL
Co-57	290-12 (at 9,400 BV)
Mn-54	1100 – 9 (at 9,400 BV)
Co-58	152 – 9 (at 9,400 BV)
Co-60	540-24 (at 9,400 BV)
Zn-65	220 – 15 ( at 19,800 BV)
Ag-110m	12-9 (at 4,700 BV)
Sb-125	41-4 ( at 4,700 BV)
Cs-134	5000-700 (at 19,800 BV)
DIABLO CANYON 1	4,000 BV processed (limited sample)
Mn-54	BDL
Co-58	795-650
Co-60	1260-660
Sb-125	12-7
DIABLO CANYON 2	4,000 BV processed (limited sample)
Mn-54	125-88
Co-58	250-180
Co-60	52-34
Ag-110m	5-3
Sb-125	3.4 (filtration)

Table VI. Overall DF's (filtration  $0.45 \,\mu\text{m} + \text{CoTreat/CsTreat}$  column exchange) for the different radionuclides.

### References

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