Evaluation of Radiochemical Properties of Legacy Dry Active Waste - 11477

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ABSTRACT

According to the Korean government 'Regulations of Low and Intermediate Radioactive Waste Delivery', radioactive waste should meet the disposal site Waste Acceptance Criteria (WAC). By December 2006, about 41,000 drums of Dry Active Waste (DAW) had been stored in Korea Hydro and Nuclear Power (KHNP) sites. These drums were packaged before the establishment of the WAC, so it was necessary to perform a radiochemical evaluation prior to disposal. The WAC includes limits on the concentration of chelating agent and free water, leaching rate, compression strength, void space, nuclide concentration, dose rate, etc. The pre-investigation of the sites, which was done by documents and visual inspection, requires all DAW meet the criteria including chelating agent and free water. The another evaluation was accomplished with a destructive method for the existing drums. The analysis was done by drum sampling (Stratified Random Sampling), which was conducted considering the decontamination process, the drum properties, and the date of drum generation. Forty one drums were selected as the sample for the four power plant sites. Another thirty five drums were selected to evaluate radionuclides. The results show that the average concentration of chelating agent (EDTA/NTA and Citric Acid) is 0.10 wt. % and the free water is 0. The gamma nuclide concentrations also satisfied the WAC. Considering that the samples are from the most conservative estimate, it could be inferred that all legacy DAW drums satisfy the WAC. The extra test from the same samples shows that the pure wastepaper sample has a higher concentration of chelating agent than other wastes, which was an expected result of Stratified Random Sampling.

INTRODUCTION

There are twenty nuclear power plants in operation in Korea, and about 1,600 drums of Dry Active Waste (DAW) are generated annually. KHNP conducted a radiochemical evaluation for the drums which were generated from 1978 to 2006. The number of DAW drums was 41,000 while the number of drums sampled was 41. The sampling method was by Stratified Random Sampling (SRS), which was conducted considering the decontamination process, the drum properties, and the date of drum generation.

The Waste Acceptance Criteria (WAC) in Korea includes limits on the concentration of chelating agent and free water, leaching rate, compression strength, void space, nuclide concentration, dose rate, etc. For DAW, chelating agent and nuclide concentration are the key factors for the disposal. Each criteria is presented in Table 1.

INVESTIGATION OF LEGACY DRY ACTIVE WASTE

Legacy DAW has been classified into fifteen categories. Among them, about 60% of the DAWs is combustible and remainder is noncombustible. When radioactive wastes were generated, they were dried first and then drummed in 200 liter containers. For volume reduction, the 200 liter drums are compressed by a super compactor to reduce the volume to around 50 %. Then two compacted drums are repacked into a 320 liter container.

The investigation determined that EDTA, Nitrilotriacetic Acid (NTA) and citric acid are the chelating agents.

Table 1. Waste Acceptance Criteria

Items		Criteria
Physicochemical Characteristic	Chelating agent	Over 0.1wt. %, explicitly labeling
	Free water	Less than 0.5 vol. %
	Void Space	Less than 15%
Nuclide Concentration	H-3	1.11E+6 (Bq/g)
	C-14	2,22E+6
	Co-60	3.70E+7
	Ni-59	7.40E+4
	Ni-63	1.11E+7
	Sr-90	7.40E+4
	Nb-94	1.11E+2
	Tc-99	1.11E+3
	I-129	3.70E+1
	Cs-137	1.11E+3
	Gross-alpha	3.70E+3

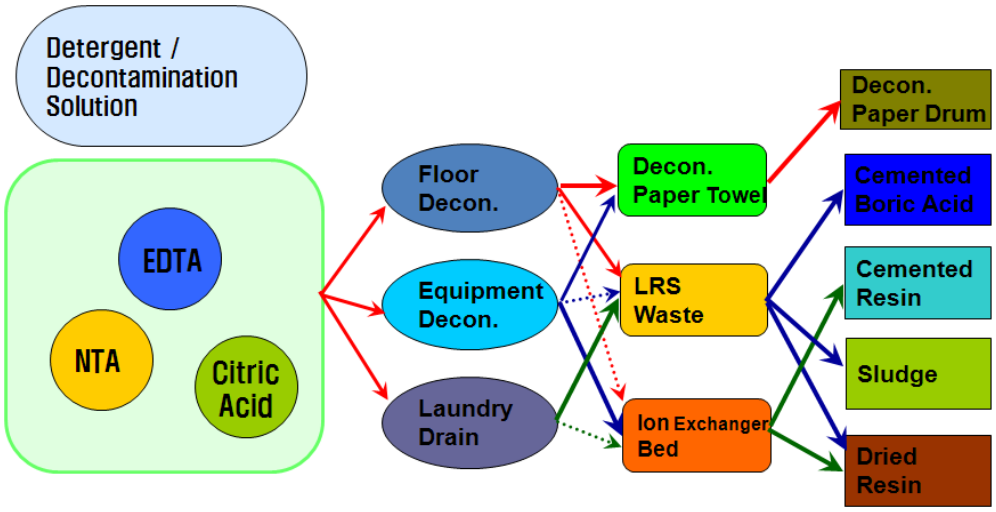

Fig. 1. Types of Chelating Agents at DAW and Their Inflow Path

It was deduced that the incoming route of the citric acid was the decontamination process of equipment, while EDTA/NTA are from decontamination of floors or other detergent processes. Figure 1 shows types of chelating agents and their typical input path.

The chelating agents contained in detergent or decontamination solution go into wastes through decontamination of floors, equipment decontamination and laundry drain. The detergent and decontamination solution used for floors would enter into dry waste via paper towels. Some of these compounds go into the Liquid Radwaste System(LRS). From this path they enter into the boric acid solidified waste, sludge and dried resin waste. The detergent and decontamination solution used for equipment decontamination enter into DAW via paper towels or spent resin waste via the liquid waste

treatment system. The laundry drain goes into the LRS only if the radioactivity is higher than discharge limit. If laundry liquid is treated by the LRS then those detergents enter into the cemented resin or the dried resin waste.

The pre-investigation of sites also shows that about 500 liters of decontamination solution containing EDTA at a level of 0.05 wt.% has been used annually per unit. This is a relatively small amount considering number of waste drums generated annually. Therefore, it is assumed that the chelate concentrations in the waste drums must be very low.

DETERMINNATION OF SAMPLE DRUMS

The number of drums sampled for destruction were minimized, but enough to be statistically valid in order to apply the analysis results to the whole set of drums. The sample drums were selected from each population of the various DAW groups. The selection was made to be one according to a Stratified Random Sampling (SRS) method the most conservative selection was conducted.

As shown in Fig. 2, the DAW was grouped into several populations according to the generation period and the maintenance type. That is, one group includes the DAW drums generated during normal operation. The other group includes the DAW drums generated during refueling outages. Each group was then divided into several populations according to the year the drum was generation. Finally, ten drums were selected randomly among each population. If there were special events that could affect the radiochemical properties of the DAW, these drums had to be included. For example, a sample drum of the forth set of the outage group(ㄴ01.11 ~ㄴ05.10) on Fig. 2 had to be included because there was decontamination work on the stud bolts during that period.

In case of the selection of drums for radionuclide analysis, special events like fuel defects and replacement of steam generators were considered. And just-before and just-after special event drums were included for the comparison.

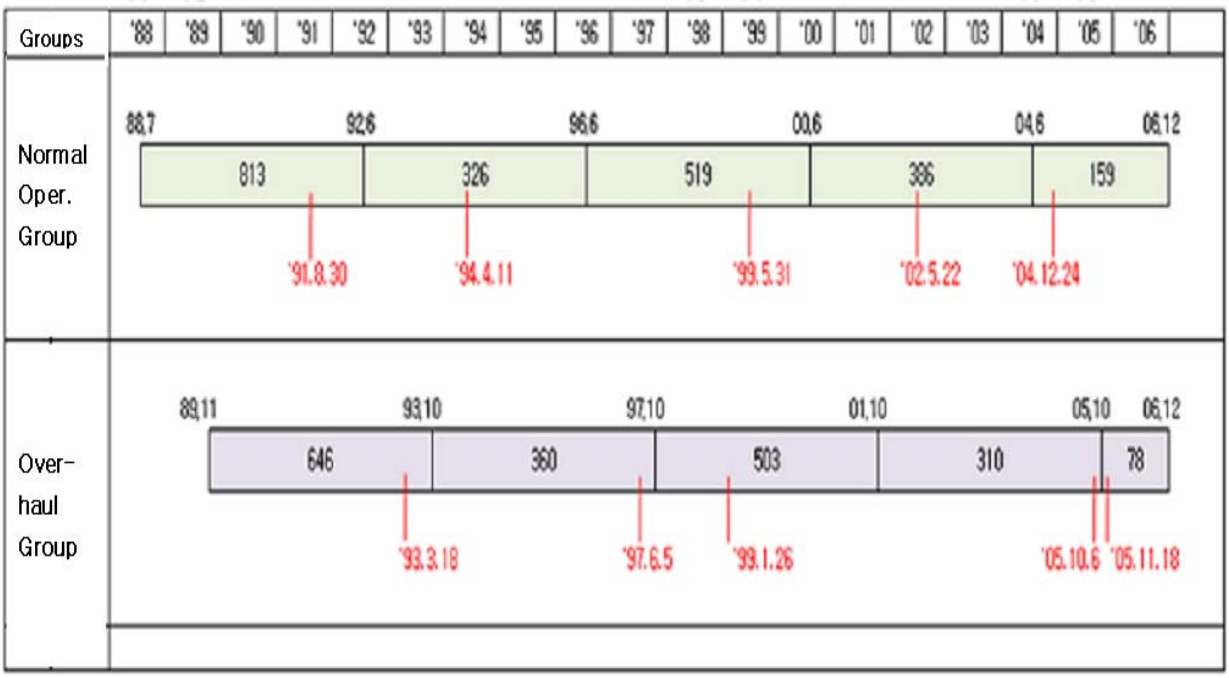

Fig. 2. Typical Sample Drums of DAW(Decontamination Papers)

RESULTS

Table 2 shows the analysis results of the DAW samples, which have been analyzed by the methods mentioned above. According to the results, the average chelate concentration of forty one samples from DAW was 0.10 wt.%. By statistical method, it can be inferred that the concentration is in the range from 0.06 wt.% to 0.14 wt.%. From the results, we found that it is difficult to say that the outage samples were higher than the normal operation samples. Figure 4 shows the picture of sample of DAW.

To evaluate the value of the decontamination paper samples, decontamination paper was solely collected from the same samples and reanalyzed. The results show that decontamination paper samples (2^{nd} analysis on the Table 3) have concentrations 1.5 times higher than those of the previous samples. (See Table 3 and Figure 3). This result proves that our assumption, that the decontamination paper samples would have the highest value, was correct.

Putting the above results together, it can be inferred that the chelate concentration of the DAW is within the guidelines of the WAC.

Table 2. Analysis Results

Items		Value
Physicochemical Characteristic	Chelating agent	0.10 wt. % (average)
	Free water	0 %
	Void Space	Less than 15%
Nuclide Concentration (calculated based on the date of the drum made)	H-3	1.4E+1 ~ 6.7E+5 (Bq/g)
	C-14	4.7E-2 ~ 4.7E+2 (Bq/g)
	Co-60	8.9E+0 ~ 1.1E+4 (Bq/g)
	Tc-99	LLD ~ 7.8E-3 (Bq/g)
	Fe-55	5.0E-1 ~ 1.1E+4 (Bq/g)
	Cs-137	9.3E-2 ~ 4.2E+1 (Bq/g)
	Gross-alpha	LLD ~ 2.0E-1 (Bq/g)

Table 3. Comparison of Chelate Concentration in Sample Group

Sample Drum		a. 1^{st} Analysis(%)	b. 2^{nd} Analysis(%)	Ratio(b/a)
Normal Operation Group	A-1991	0.03	0.03	1
	B-1994	0.02	0.12	6
	C-1999	0.06	0.22	3.6
	D-2002	0.05	0.08	1.3
	E-2004	0.15	0.21	1.4
Overhaul Group	F-1993	0.03	0.03	1
	G-1997	0.30	0.24	0.8
	H-1999	0.02	0.08	4
	I-2005	0.02	0.03	1.5
	J-2005	0.05	0.08	1.6
	Average	0.07	0.11	1.57

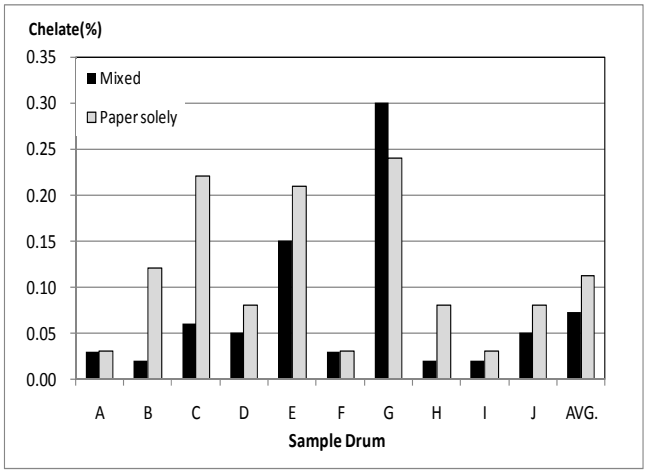

Fig. 3. Comparison of Chelate Concentration in Sample

Fig. 4. Picture of Sample of DAW

CONCLUSION

For the characterization of the legacy DAW in Korea, forty one samples have been selected and analyzed. The results show that the average chelate concentration was 0.10 wt.%, which is induced statistically in the range of 0.06 wt.% to 0.14 wt.%.

To evaluate the validity of the analyzed results of decontamination paper, decontamination paper was collected solely from the same samples and analyzed again. The results show that decontamination paper samples had 1.5 times (maximum 3.6 times) higher concentrations. Because these results are for the decontamination paper only in DAW, which was expected to be the most conservative set, it can be inferred that the DAW will satisfy the disposal criteria requested by the Korean regulatory body.

REFERENCES

1. Acceptance Criteria for Low- and Intermediate-Level Radioactive Waste, Notice of the Minister of Education, Science and Technology No 2009-37 (2009), MEST
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