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RESEARCH AND DEVELOPMENT ON TREATMENT OF LIQUID RADIOACTIVE WASTES IN THAILAND

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Abstract

The studies have been directed towards treatment technologies for low level waste. The simple physico-chemical method has been studied for applying to various kinds of waste streams such as reactor waste, isotope production waste and liquid waste from the hospitals. The characterization of inorganic ion exchangers including the effect of pH, equilibrium time, temperature and concentration of such exchangers were tested. The results revealed that the local simple brand-washed detergents, which are very cheap, can be succesfully used for decontamination instead of a more expensive imported decontaminating agent. It was also revealed that chemical precipitation can be successfully used forthe treatment of such wastes.

In considering an immobilization process for the treated waste, cementation was selected. The basic properties of the cemented waste forms have been investigated including leachability of the cemented sludge resulted from the chemical precipitation of the decontamination waste. The results revealed that the cemented inorganic ion exchangers and the sludge waste exhibit high compressive strength and low leach rates. The compressive strength of 118-207 kg/cm^2 and 15% and 20% waste loading was found to be optimum for the waste forms. A cumulative fraction leached rate from the cemented sludge was found to be about 30 x 10^{-3} cm/day at 30 day leaching time.

1. INTRODUCTION

The application of radionuclides in research, medicine, agriculture and industry in Thailand has been gradually increased since the introduction of the "Atom for Peace" programme in Thailand in 1962. A wide range of applications generates a variety of radioactive aqueous waste streams needed to be treated prior to release to the environment. The Office of Atomic Energy for Peace (OAEP) has been assigned to take the responsibility for the safe management of such radioactive wastes. All radioactive wastes generated by any user in Thailand are sent to the OAEP for treatment and subsequent storage and disposal. A survey of sources, types, volumes and characteristics of radioactive wastes arising in Thailand has been carried out. The information obtained from the survey was used for design of a new waste treatment plant to be in 1997. The data are summarized in Table I.

2. TREATMENT OF LOW-LEVEL RADIOACTIVE WASTE BY INORGANIC ION EXCHANGERS

The study was aimed at selecting inorganic ion exchangers that are inexpensive and suitable for conditioning and disposal. The experiments were conducted to determine the sorption characteristics of various inorganic ion exchangers. The sorption capacity of the ion exchangers were tested for ^{137}Cs and ^{99}Tc under various conditions including the effect of pH, equilibrium time, temperature and concentration (weight). Bentonite, kaolinite clay, sand and sandy soil were used as natural inorganic exchangers. Titanium dioxide, zeolite, antimony pentoxide and hydrated antimony pentoxide (HAP) were used as synthetic inorganic exchangers. Furthermore a basic study on the cementation immobilization process was conducted and basic properties of waste forms such as physical stability, compressive strength and leachablility were tested.

TABLE 1. DISTRIBUTION OF LIQUID WASTE STREAMS

Waste stream	%	Major radionuclides	Composition
Medical	32	^{67}Ga, ^{51}Cr, ^{99m}Tc, ^{131}I ^{201}Tl, ^{3}H, ^{14}C	Laboratory and decontamination wastes Pump oil, scintillators
Education/ research	43	^{32}P, ^{35}S, ^{51}Cr, ^{45}Ca, ^{99m}Tc, ^{131}I, ^{60}Co, ^{137}Cs	Liquids, hot cell experiments Serum blood
Reactor operations	20	Fission and activated corrosion products	Reactor coolant, storage pool water
Monazite extraction	5	U, Th and their daughter products	Extraction solvent, plant wastes

Total waste volume: aqueous solutions = 200 - 300 m^3, organic waste = 6 - 8 m^3 per year.

It was found that the sorption capacity of titanium dioxide, zeolite, bentonite, kaolinite, montmorillonite, sand, sandy-soil, hydrated antimony pentoxide (HAP) and antimony pentoxide for ^{137}Cs were 70, 99, 80, 98, 98, 86, 87, 85 and 98%, respectively. Optimum conditions for the sorption of ^{137}Cs on the exchangers are listed in the Table II.

TABLE II. THE OPTIMUM SORBTION CONDITIONS FOR ^{137}Cs

Exchangers	pH	Time (min)	Concentration (g/mL)	Temperature (°C)
Titanium dioxide	3-6	20	3	25-50
Zeolite	1-13	5	0.3	25-50
Bentonite	1-13	20	1	25-50
Kaolinite	3-11	10	0.3	25-50
Montmorillonite	3-10	15	0.2	25-30
Sand	3-11	20	1	25-30
Sandy soil	3-11	20	1	25-30
HAP	1-7	30	1	25-30
Antimony peroxide	1-9	20	1	

* The volume of test solution is 30 mL each.

The sorption capacity for ^{99m}Tc on all but one of the inorganic ion-exchangers under investigation was very poor. Only the antimony pentoxide specimen has shown a good sorption capacity of about 80-90%. The optimum condition was found at pH1-9, the optimum time of 5 days and weight-ratio of 3 g to 30 mL of solution.

3. TREATMENT OF LOW LEVEL RADIOACTIVE WASTE BY CHEMICAL PRECIPITATION

3.1. CHEMICAL TREATMENT OF LIQUID WASTE FROM THE CHEMICAL EXTRACTION OF MONAZITE

The studies were performed to find the best condition for the treatment of a waste stream from the monazite extraction laboratory which contains a very low level of U/Th and their daughter products. The parameters included the type and the concentration of the chemical coagulant, efficiency of treatment, pH, rate of sedimentation including the coagulant aids. The results obtained as shown in Table III, disclosed that the aluminium and the barium chloride were main chemical coagulants for the treatment of monazite extraction process waste and could give a decontamination factor up to 20.

TABLE III. OPTIMUM CONDITIONS FOR THE CHEMICAL TREATMENT OF THE LIQUID WASTE STREAM FROM THE MONAZITE EXTRACTION PROCESS

Chemicals	Concentration (ppm)	pH before	pH after	Removal (%)	DF
Alum NaOH $BaCl_2$ Orgatite Separan	200 600 700 6 10	3.85	8.9	95.53	22.41
Alum Na_2CO_3 $BaCl_2$ Orgatite Separan	200 800 700 6 12	3.85	5.6	88.26	8.52
Alum $Ca(OH)_2$ $BaCl_2$ Orgatite Separan	200 500 700 8 8	3.85	7.4	95.52	21.07

3.2. CHEMICAL TREATMENT OF THE DECONTAMINATION AND LAUNDRY WASTE

In a radiation laboratory, it is necessary to clean the items contaminated with radionuclides every day. This process creates low level liquid wastes of various types and quantities. An attempt to minimize a number of types of the waste from such cleaning process has been made. One of the actions were to encourage the radioisotope users to use only a few branded names of detergents and decontaminating agents. To achieve such a goal, R&D for an appropriated detergent and decontaminating agent has been commenced.

3.2.1. Detergent efficiency in the radioactive material decontamination

Searching of the most suitable detergents was undertaken by counting the activity of the fixed-form contaminated radioactive tracers on 3 kinds of cloth which were cotton, calio and polyester, before and after being washed by the 24 locally available commercial detergents. The washing efficiency of 24 detergents for radioactive decontamination of cotton, calio and polyester varied from 0-98.47% and can be summarized as follows:

Hand-washed powder detergent:	*cotton > calio > polyester*
Machine-washed powder detergent:	*polyester > cotton > calio*
Hand and machine-washed liquid detergent:	*calio > polyester > cotton*
Pre-washed stain remover:	*calio > cotton > polyester*
Commercial decontaminating liquid:	*cotton > calio > polyester*

3.2.2. Treatment of the waste from laundry and decontamination processes

A machine-washed liquid detergent was selected to be used as a detergent and a decontamination agent in a radiochemical laboratory. The composition of the detergent was as follows:

Detergent composition

Dodecycle benzene sulfonic acid sodium (DBS)	12.5%
Sodium tripolyphosphate	25.0%
Nitrilotriacetic acid	30.0%
Sodium sulphate, anhydrous	25.0%
Citric acid	6.25%
Carboxymethyl cellulose	1.25%

The waste from the washing and decontaminating processes carried out in the Office of Atomic Energy for Peace Laboratory were collected and characterized. Characteristics of the waste stream was as follows:

Characteristics of the waste water after the washing process

pH	7.21± 0.01	
Ca^{+2}	19.03±0.48	ppm
K^{+}	2.61±0.01	ppm
NO_3^-	3.77±0.4	ppm
Cl^-	25.0±1.30	ppm
PO_4^{-3}	> 10	ppm.
Temperature	24.83±0.04	°C
Conductivity	418.75±1.30	ms/cm
Total dissolved solid	281.75±0.83	ppm
Total hardness	86.25±0.83	ppm
Dissolved oxygen	2.28± 0.04	ppm
Activity		
gross alpha	< 1.02 x10^{-5}	Bq/L
gross beta	0.145±0.002	Bq/L

Coagulation-precipitation methods have been tested to find the best condition for the treatment of the waste stream. The results obtained are shown in Table IV.

TABLE IV. THE PRECIPITATION CHARACTERISTICS FOR THE WASHING WASTE

Precipitation method	Chemicals	pH	Radionuclide	%Removal
Phosphate coagulation	$Na_3PO_4.12H_2O$ $Ca(OH)_2$	10	^{90}Sr	93.3
Copper Ferrocyanide coagulation	$K_4Fe(CN)_6.5H_2O$ $CuSO_4.5H_2O$ Bentonite	5-6	^{134}Cs	100
Cobalt precipitation	$Fe(NO_3)_3$ $.9H_2O$ NaOH Diethyl dithio-sodium carbamate	10-11	^{60}Co	100

4. IMMOBILIZATION OF THE TREATED WASTES

Immobilization of radioactive waste is a step required to produce a waste form suitable for handling, transportation, storage and disposal. Immobilization processes involve conversion of waste to solid forms that reduce the potential for migration or dispersion of radionuclides from the waste. The cementation method was chosen for the studies on the immobilization of ion exchange materials as mentioned in Section 3.1 and the sludge from the washing process as mentioned in Section 3.2.

4.1. THE BASIC PROPERTIES OF THE CEMENTED WASTE FORMS

Physical and mechanical properties of the cemented waste form have been investigated. The results are given in Tables V and VI.

TABLE V. DENSITY OF THE CEMENTED ION-EXCHANGE MATERIALS

Ion exchange materials	Density (g/cm^3)
Titanium dioxide	0.7056
Zeolite	0.5967
Bentonite	0.7954
Kaolinite	0.9716
Sand	1.3667
HAP	0.8392
Antimony pentoxide	1.6579
Portland cement (without additive)	1.3581

(* Normal Portland cement was used in this study)

TABLE VI. OPTIMUM WEIGHT RATIO OF THE ION EXCHANGER AND THE SLUDGE TO CEMENT

Ion exchanger materials	Weight ratio	Compressive strength (kg/cm^2)
Titanium dioxide	45:55	128-148
Zeolite	25:75	188-207
Bentonite	19:81	118-138
Kaolinite	24:76	128-147
Sand	54:45	180-201
HAP	--	--
Antimony pentoxide	--	--
15% loading of sludge	*34:50*	*160-166*
20% loading of sludge	*32: 47*	*150-160*

4.2. LEACHABILITY

Leachability of the simulated waste forms was measured using the Brookhaven National Laboratory's Accelerated Leach Test Method. The leaching rate (LR) and the cumulative leached fraction (CLF) are defined as follows:

$$\text{Leach rate (LR)} = (A_t \times W_o)/(A_o \times S \times t)\ g/cm^2.d$$

where

W_o is the initial weight of the sample,
S is the surface area of the sample (cm^2), this experiment~83.8 cm^2,
A_o is the initial activities in the cemented sample,
A_t is the activity in the leachant removed in time (t), and
t is the leaching time (day).

Leach factor (LF) = $\sum a \cdot V/A_o \cdot S$ cm/t

where

V is the volume of the specimen (cm^3),
S is the surface area of the specimen (cm^2), and
$\sum a$ is the total amount of the radionuclide leached out in t leaching interval.

The results of the leaching test for 15% and 20% loaded sludge are shown in Table VII.

TABLE VII. LEACHABILITY OF THE CEMENTED WASTE FORMS

Leaching time(d)	CLF x 10^{-3} (cm/day) 15% loaded sludge at 25°C	CLF x 10^{-3} (cm/day) 15% loaded sludge at 50°C	CLF x 10^{-3} (cm/day) 20% loaded sludge 25°C	CLF x 10^{-3} (cm/day) 20% loaded sludge 50°C
0.08	3.13	8.97	3.14	17.54
0.2	8.77	17.55	8.77	21.04
1	11.22	21.05	11.23	23.42
3	14.21	25.47	14.22	25.46
7	15.67	27.91	15.67	27.91
10	16.27	28.78	16.27	28.78
14	16.73	29.56	16.73	29.56
21	17.10	30.15	17.10	30.17
30	17.37	3 0.61	17.37	3 0.61

5. CONCLUSION

The simple method has been studied for applying to many kind of waste streams such as reactor waste, isotope production waste and liquid waste from hospitals. It is concluded that the inorganic ion exchangers can be used for the treatment of the low-level liquid wastes. But for implementation on a larger scale, there is a need to consider how to control the optimum condition.

The results from the investigation on the efficiency of detergent in the radioactive decontamination revealed that the locally available hand and machined-washed detergents, which are very cheap, can be used as a decontaminating agent instead of the more expensive imported decontaminating agents. According to the results received, the OAEP will recommend the radioisotope laboratories to use the simple detergent for radioactive work, so that the waste stream from the waste generators will contain a similar composition of laundry waste. The treatment method for such a waste as indicated in the investigation revealed that chemical precipitation methods can be used successfully for treating of such a waste arising.

Considering the immobilizing process for treated waste, cementation method was selected. The basic properties of the cemented waste forms have been investigated as well the leachability of a particular type of sludge. The result revealed that cemented waste forms of inorganic ion exchangers and sludge from the chemical precipitation of decontamination waste exhibited a high compressive strength and a low leach rate.

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