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REMOVAL OF ZN-65, MO-99 and I-125 FROM EFFLUENT BY COAGULATION-FLOCCULATION PROCESS

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ABSTRACT

The aim of this study is to investigate the efficiency treatment in removing Zn-65, Mo-99 and I-125 from an aqueous radioactive effluent. The wastes are currently being produced from hospitals, research institutes, clinics and universities. Effluent was spiked separately with each type of the radioisotope and was treated by the coagulation-flocculation process. By varying the chemical dosages (i.e., alum, soda ash, ferric chloride and coagulant aid) in the treatment, different decontamination factor values were obtained. Optimum dosages and types of chemical used to remove a particular radioisotope was determined. Results indicated that optimum pH value for removing Zn-65 in an effluent was pH 8. The highest decontamination factor value was 61. In removal of I-125 radioisotope, ferric chloride was suitable as a coagulant that gives the highest decontamination factor value of 5.0. Treatment to remove Mo-99 radioisotopes was conducted in the laboratory and treatment plant scale. For Mo-99 radioisotope treatment by laboratory and plant scale, the highest decontamination factor obtained was between pH values of 4.0 to 4.5. By extrapolation of both scales, the plant scale treatment does not vary significantly from laboratory scale. This indicated treatment dosages of chemicals for the Low Level Treatment Plant scale be deduced from the laboratory scale.

INTRODUCTION

Malaysian Institute for Nuclear Technology Research (MINT) has been receiving aqueous radioactive wastes containing Zn-65, Mo-99 and I-125 from hospitals, medical clinics, research institutes and universities. Table 1 shows the characteristic of the wastes being produced. The radioisotopes have toxic properties and with half-life between 2.75 to 59.7 days can pose an environmental hazard if the effluent is not properly treated before release to the environment[1].

Coagulation flocculation process can treat effluent containing radioactive material. Release of treated effluent must fulfill the conditions of the Environmental Act 1974 of Malaysia where the pH range between 5.5 to 9.0, COD < 50 mg/l, BOD < 20 mg /l and suspended solid < 50 mg/l. According to recommendation by IAEA, release of 20 m^3/week of radioactive liquid wastes to the environment, the activity must be less than 10^{-5} μCi/ml[2].. This limit is based on the liquid wastes that contains Sr-90. Water quality parameter values of the effluent must be adjusted to the permissible values before being release to the environment [3-4].

Main mechanisms for removing radionuclides in the coagulation-flocculation process are [5-6]: 1) Direct flocculation and coagulation process, 2) Precipitation and co-precipitation, 3) Adsorption on the coagulant aid, 4) Ion-exchange mechanism, and 5) Physical enmeshment by coagulant aid. For a given effluent water there is at least one pH range that contributes a good coagulation-flocculation process in the shortest time with a given chemical coagulant dose [7-8].

OBJECTIVE OF EXPERIMENT

By Jar Test method, studies were conducted to determine the efficiency treatment to treat effluent wastes containing Zn-65, Mo-99 and I-125 [9-10]. Type and suitable chemical dosages were also identified at the required optimum conditions to reduced as much effluent radioactivity in the treatment.

MATERIALS AND METHOD

Treatment of Effluent Containing Zn-65

The influence of pH on radioactivity removal has been studied by varying pH between range of 4.5 to 8.5. This was done by varying the soda ash dosage but maintaining the alum and coagulant aid dosage constant. The volume of sample tested was 1000 ml. Blank sample was prepared containing alum, soda ash and coagulant aid but without Zn-65 radionuclide. Gas proportional counter (Canberra Model 2400) was used to count samples activities for 20 minutes before and after treatment [11]. Optimum pH range for maximum Zn-65 removal was determined from the percentage of radionuclide removed or from the decontamination factor values.

There is an optimum alum dosage when the treatment condition was maintained at the optimum pH value. Good estimation of soda ash and alum dosage were used to maintain the optimum pH condition. Optimum alum dosage was determined by the highest percentage of Zn-65 radionuclide removed from the effluent.

Treatment of Effluent Containing Mo-99 at Laboratory and Plant Scale

Both laboratory and plant scales were performed to treat effluent containing Mo-99 radionuclide. The alum and coagulant aid dosage were maintained while the soda ash dosage was gradually increased. Chemicals' dosage used for each batch treatment of 1600 liters effluent in plant scale was derived by ratio from each laboratory scale treatment for 1 liter effluent. Percentage of radionuclide removed at different pH values at laboratory and plant scale was compared to determine any large differences in removal of Mo-99 radioisotope. Proportional gas counter (Canberra Model 2400) was used to count the net beta activity for each representative sample. All samples' activities were corrected for decay before treatment because of the short half life of Mo-99 (2.75 days).

Treatment of Effluent Containing I-125

In first treatment process, alum, soda ash and coagulant aid (Praestol) were used to treat spiked I-125 effluent wastes. I-125 in the form of NaI in sodium solution from Amersham International, U.K. was used in the experiment. The spiked effluent activity was diluted to about the same effluent activity received from users. Proportional gas counter was used for counting of the samples. The same dosages of alum (4 ml of 100 g/L concentration) and Praestol (4 ml of 0.25 g/L concentration) were used, while dosages of soda ash (100 g/L) were varied with an increment of 1 ml.

In second treatment process, the same dosages of ferric chloride (5 ml of 100 g/L concentration) and Praestol (4 ml of 0.25 g/L concentration) were used. The dosages of soda ash used were varied in order to have sol ution at the required pH condition.

Formulas Used in Calculation

$$A = A_o \exp((-0.693 \times t)/T) \qquad (1)$$

where, A = Activity after time t, cpm
Ao = Initial activity before treatment at to, cpm
t = Time taken (t-to), sec
T = Half life, sec
x = Multiplication sign

$$\text{Sample activity} = \frac{(A-B) \times 1000 \times 1}{E \times V \times Min \times 60} \text{ Bq/l} \qquad (2)$$

where, A = Average reading for sample
B = Average reading for background
E = Efficiency of detector (34% for Canberra Model 2400)
V = Sample volume (ml)
Min = Counting time (minute)

$$\%\text{Radionuclide Removed} = \frac{(\text{Original Activity - Final Activity}) \times 100}{\text{Original Activity}} \quad (3)$$

$$\text{Decontamination Factor (DF)} = \frac{\text{Activity of Effluent Before Treatment}}{\text{Activity of Effluent After Treatment}} \quad (4)$$

RESULTS AND DISCUSSION

Zinc-65 Treatment

Table 2 shows the variations of decontamination factor and percentage activity removed at different pH values. Figure 1 shows that optimum pH value was 8 corresponding to the highest decontamination factor value for removal of Zn-65. Figure 1 also shows percentage radionuclide removed after pH 8 when the curve indicated there was a decrease in percentage of radionuclide removed.

It was observed that alum and soda ash can affect the size of particles produced. Reduction of particle size was produced when the dosage of soda ash decreased. This caused lesser precipitation resulting smaller decontamination factor value. When soda ash dosage increased, there was an increased in colloid particles produced.

Treatment data to determine optimum alum dosage is shown in Table 3. From Figure 2, optimum alum dosage was 7 ml when percentage of radionuclide removed was nearly optimum at 95 %. Major portion of the Zn-65 radionuclide was removed in the effluent.

Mo-99 Treatment

Treatment results at laboratory and plant scale are shown in Table 4 and 5, respectively. From Figure 3, pH range for removing about 80% of Mo-99 is between 4 and 4.5. There was small difference along the two curves for the two scales' treatments. The results show that the Jar Test experiment at laboratory scale are valid for extrapolation to the plant scale treatment.

Similar condition, methods and chemicals determined by laboratory scale treatment can be used, after adjustment accordingly, for effluent treatment in the plant. The amount of chemicals for plant scale treatment can be calculated by ratio from the chemical's dosage used at laboratory scale. Treated effluent pH which has to be below the permissible discharge limit pH 6 to pH 9, can be adjusted by the addition of Soda Ash.

I-125 Treatment

Table 6 and 7 show results using ferric chloride and alum as coagulants. From Figure 4, when alum was used the highest decontamination factor value was 2 in the lower region between pH 5 to 6. The decontamination factor values decreased between pH 6 to 8 and slightly increased again after pH 8. The pH condition range between 5 to 6 was suitable in this effluent treatment.

From Figure 5, ferric chloride as coagulant, decontamination factor value of 5 was obtained between pH 6 to 7. The decontamination factor value decreases steadily after pH value 7.

From the two experiments conducted, ferric chloride contributed higher decontamination factor value than alum as coagulant. This indicated that ferric chloride was suitable for the treatment of I-125 effluent.

CONCLUSION

From this study, for a given liquid effluent there is at least one pH range that contributes a good coagulation-flocculation process with a given chemical coagulant dose. Experiments conducted show that removal of Zn-65 was optimum at pH value 8 with decontamination factor value of 61. Optimum alum dosage for removing Zn-65 at the pH 8 was 7 ml. For Mo-99 radioisotope treatment by laboratory and plant scale, the highest decontamination factor value determined was between pH range of 4.0 to 4.5. Extrapolation of the laboratory scale shows that the plant scale treatment does not vary significantly. The procedure conducted is valid for the conditions, methods and chemicals from laboratory scale treatment to be adjusted accordingly for plant scale treatment. For I-125 treatment, ferric chloride was more suitable than alum as a coagulant which produced higher decontamination factor value 5 at pH 7 condition.

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TABLE 1. Zn-65, Mo-99 and I-125 Aqueous Wastes Characteristics Received From Producers

Radioisotope	Source of Wastes	Volume (Liter)	Concentration KBq/l
Zn-65	Universities, MINT, Research Institutes	60	4.0
Mo-99	MINT	15	40
I-125	General Hospital, Research Institutes	6938	57.0

TABLE 2. Soda Ash Dosage Change with Alum (4 ml) and Coagulant Aid Dosage (1 ml) at Constant Volume

Test No.	Soda Ash (ml)	pH	Average Act. (cpm)	Net Act. (cpm)	Final Act. (cpm)	Decontamination Factor (DF)	% Radionuclide Lost
Blank	4.0		6748				
1	3.0	4.5	7836	1088	989	1.1	9.0
2	4.0	5.6	7762	1014	859	1.2	15.3
3	5.0	6.0	7266	518	224	2.3	56.7
4	7.0	6.9	6959	211	30	7.0	85.8
5	8.0	7.1	6810	62	9	6.9	85.5
6	9.0	7.6	6831	83	6	13.8	92.8
7	10.0	8.0	6768	20	0.3	60.6	98.4
8	11.0	8.5	6863	115	11	10.5	90.4
9	15.0	9.3	6857	109	9.9	11.0	90.9

TABLE 3. Determination of Optimum Alum Dosage with Coagulant Aid Dosage (1 ml) at Constant pH Value 8.

Test No.	Soda Ash (ml)	Alum (ml)	Ave. Act. (cpm)	Net Act. (cpm)	Final Act. (cpm)	Decontamination Factor (DF)	% Radionuclide Lost
Blank	4.0	4.0	3338				
1	2.5	1.0	4333	995	169	6.0	83
2	5.0	2.0	3418	80	11	7.0	86
3	7.5	3.0	3411	73	10	7.5	87
4	10.0	4.0	3385	47	4	12.0	92
5	12.5	5.0	3707	69	9	8.1	88
6	13.0	5.5	3372	34	2	16.5	94
7	14.0	6.0	3354	16	0.5	35.6	97
8	19.5	8.0	3354	16	0.5	35.5	97
9	24.0	10.0	3417	79	11	7.0	86
10	29.0	12.0	3361	23	0.9	24.5	96

TABLE 4. Laboratory Scale Treatment with Alum (4 ml) and Coagulant Aid (1 ml) at Constant Volume

Test No.	Final pH	Average Net Activity (cpm)	Final Activity (cpm)	Bq/l	% Radionuclide Lost
1	4.5	3508	829	406	76
2	4.5	3508	1090	464	69
3	5.5	3508	1727	847	51
4	5.9	3508	2570	1260	27
5	6.2	3508	2802	1373	20
6	6.5	3508	3100	1520	12
7	6.7	3508	3461	1692	1

TABLE 5. Plant Scale Treatment

Test No.	Final pH	Average Net Activity (cpm)	Final Activity (cpm)	Bq/l	% Radionuclide Lost
1	3.9	3508	875	428	75
2	5.7	3508	1954	954	44
3	6.1	3508	2604	1277	26
4	6.3	3508	2927	1435	17
5	6.5	3508	2858	1401	19
6	7.0	3508	3219	1578	8
7	7.2	3508	3207	1572	9
8	7.8	3508	3219	1578	8
9	6.9	3508	3020	1480	14
10	4.5	3508	691	339	80

TABLE 6. Results of Effluent Treatment Using 4 ml of 100 g/L Alum as Coagulant and 4 ml of 0.25 g/L of Coagulant Aid

Sample	Soda Ash (ml)	pH	Conductivity (µS)	Counts Per Min (cpm)	Decontamination. Factor (DF)
Spiked Sol. (Untreated)	-	7.2	176	89	-
1	1.0	4.9	405	46	1.9
2	2.0	5.9	483	47	1.9
3	3.0	6.2	572	51	1.8
4	4.0	6.7	727	68	1.3
5	5.0	6.4	808	66	1.4
6	6.0	7.0	978	73	1.2
7	7.0	7.5	1093	79	1.1
8	8.0	8.3	1228	78	1.1
9	9.0	8.5	1306	68	1.3
10	10.0	8.5	1286	59	1.5

Table 7. Results of Effluent Treatment Using 5 ml of 100 g/L of Ferric Chloride as Coagulant and 4 ml of 0.25 g/L of Coagulant Aid

Sample	Soda Ash (ml)	pH	Conductivity (µS)	Counts per min. (cpm)	Decontamination Factor (DF)
Spiked sol. Untreated	-	8.0	208	115	-
1	3.0	6.1	727	25	4.6
2	5.0	7.0	890	24	4.8
3	9.5	8.0	1351	33	3.4
4	11.0	9.0	1496	41	2.8
5	19.5	10.0	2310	51	2.3

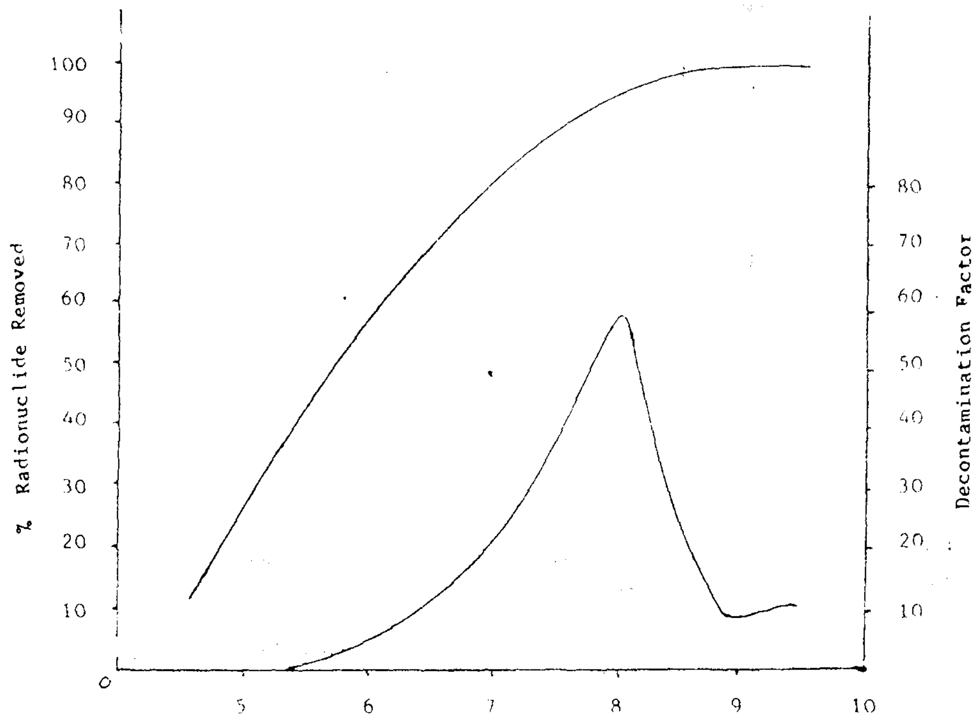

FIGURE 1. Graph % Radionuclide Removed against pH and Decontamination Factor against pH

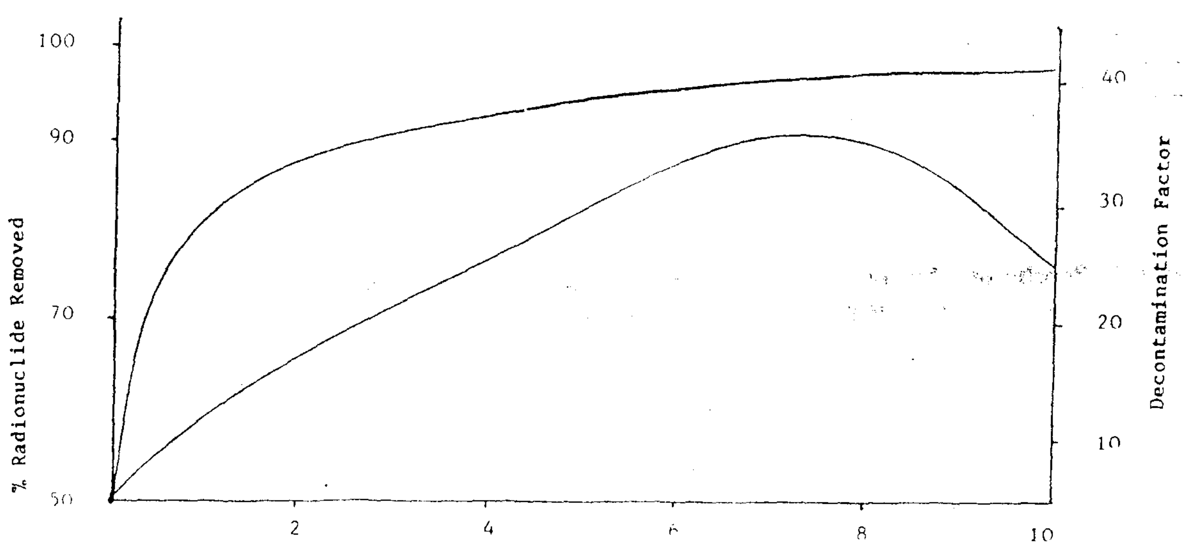

FIGURE 2. Graph % radionuclide Removed against Alum (ml) and Decontamination Factor against Alum (ml)

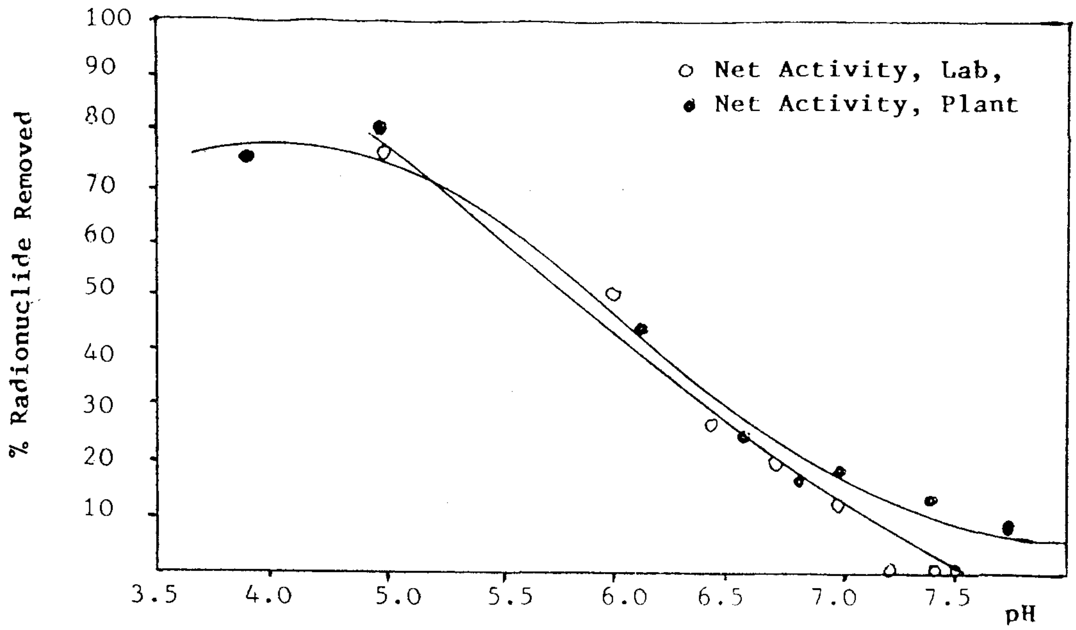

FIGURE 3. Comparison of Treatment in Laboratory and Low Treatment Plant

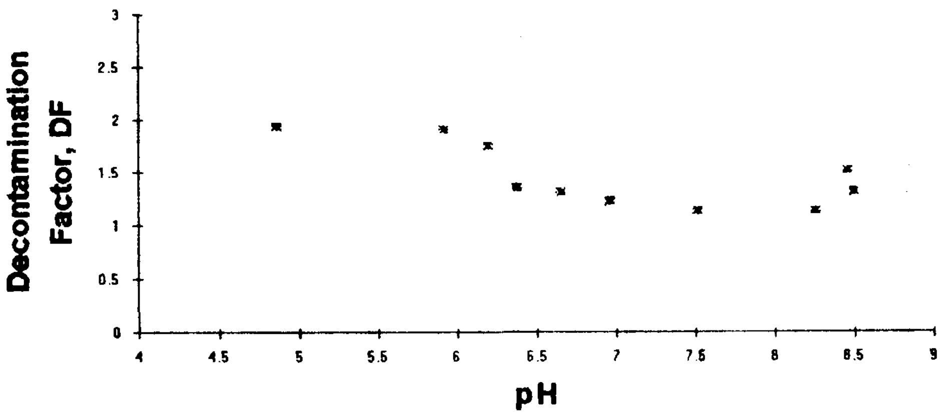

FIGURE 4. Chemical Treatment Alum as Coagulant

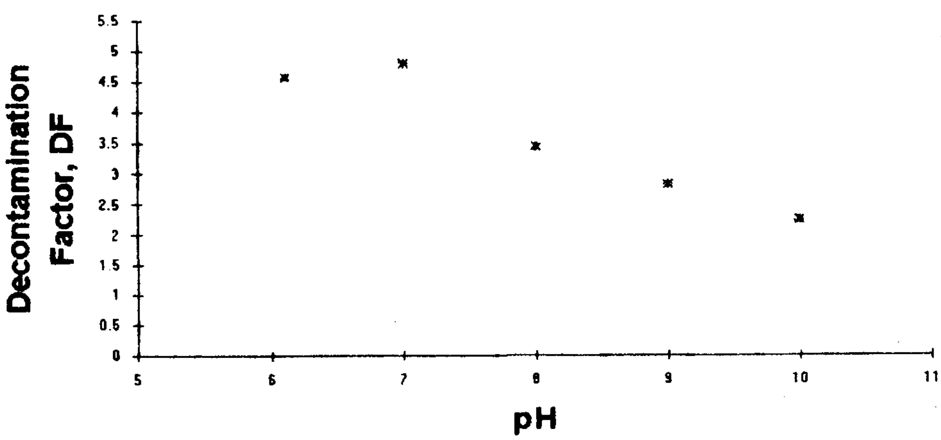

FIGURE 5. Chemical Treatment Ferric Chloride as Coagulant