DEVELOPMENT OF IMPROVED RADIOACTIVE EFFLUENT TREATMENT TO REMOVE Zn-65, Mo-99 AND I-125 BY THE COAGULATION-FLOCCULATION PROCESS

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Abstract

Coagulation-flocculation treatment using aluminum sulphate, sodium carbonate, ferric chloride and coagulant aid was able to remove ^{65}Zn, ^{99}Mo and ^{125}I from an aqueous effluent. Chemicals' dosages into the samples were varied which contributed different decontamination factors. For ^{65}Zn removal, optimum pH value was 8 that provided the decontamination factor of 35. For ^{125}I, optimum pH value was 7 with the decontamination factor of 4.8. Treatment of the effluent containing ^{99}Mo at a laboratory scale was proved to be valid for the extrapolation to a plant scale. The pH range for optimum treatment was between 4.0 to 4.5.

1. INTRODUCTION

Treatment of effluents containing radioactive material can be conducted by using flocculation-coagulation process [1-4]. Main mechanisms for removing radionuclides in the process are: (1) Directly from coagulation and flocculation process, (2) Precipitation and co-precipitation, (3) Adsorption on the coagulant aid, (4) Ion-exchange and (5) Physical enmeshment by coagulant aid.

The treated effluents to be released to the environment must fulfill the conditions of the Environmental Act 1974 of Malaysia where the pH ranges between 5.5 to 9.0, COD < 50 $mg \cdot L^{-1}$, BOD < 20 $mg \cdot L^{-1}$ and suspended solid < 50 $mg \cdot L^{-1}$. For the release rate of 20 m^3 of radioactive liquid wastes per week, their activity must be less than 10^{-5} $\mu Ci \cdot mL^{-1}$ [5]. This limit is established for the liquid wastes contaminated with ^{90}Sr.

The aim of the study was to improve treatment of radioactive effluents to reduce the content ^{65}Zn, ^{99}Mo and ^{125}I as low as possible by the coagulation-flocculation process.

2. EXPERIMENTAL METHODS

For a given radioactive effluent, there were the interrelated optimum conditions for treatment. The conditions included pH, turbidity, chemical composition of the water, type of coagulant as well as physical factors such as temperature and mixing conditions. Jar test experiments were conducted using different dosages of chemicals for each treatment of the effluent sample containing radionuclides ^{65}Zn, ^{99}Mo and ^{125}I [1-4]. In the coagulation-flocculation process there was at least one pH range that provided the best results in the shortest time with a given chemical coagulant dosage. The volume of each sample test conducted was 1000 mL.

2.1. TREATMENT OF THE EFFLUENT CONTAINING ^{65}Zn

pH values of the effluents varied within the range 4.5-8.5. The adjustment was done by varying the soda ash dosage and maintaining the alum and coagulant aid dosages constant. A blank sample without ^{65}Zn radionuclide was also prepared which contains alum, soda ash and coagulant aid. A gas proportional counter (Canberra Model 2400) [6] was used to count the samples' activities for 20 minutes before and after treatment. In another experiment, an optimum alum dosage was determined at the optimum pH condition. Good estimation of the soda ash and alum dosages was used to maintain optimum pH condition.

2.2. TREATMENT OF THE EFFLUENTS CONTAINING ^{99}Mo AT LABORATORY AND PLANT SCALES

Both laboratory and plant scale experiments were conducted to treat the effluents containing ^{99}Mo. The alum and coagulant aid dosages were maintained while the soda ash dosage was gradually increased. The chemicals dosage used for each batch treatment for 1600 L effluents in the plant was calculated by ratio from 1 L effluent treatment on a laboratory scale. The percentages of the radionuclide removed at different pH values at a laboratory scale and at a plant scale were compared to determine any large differences in removal of ^{99}Mo. The proportional gas counter (Canberra Model 2400) was used to count the activity from each sample. All samples' activities obtained from both treatment methods were corrected for decay before the treatment because of the short half-life of ^{99}Mo (2.75 days).

2.3. TREATMENT OF THE EFFLUENTS CONTAINING ^{125}I

Two jar test experiments were conducted each using different chemicals. The experimental conditions for the two tests were similar. The same dosages of alum (4 mL of 100 g·L^{-1} concentration) and coagulant aid Praestol (4 mL of 0.25 g·L^{-1} concentration were used in the first test. The dosages of soda ash (100 g·L^{-1}) were varied with an increment of 1 mL. In the second test, the same dosages of ferric chloride (5 mL of 100 g·L^{-1} concentration) and Praestol (4 mL of 0.25 g·L^{-1} concentration were used. The dosage of soda ash was varied according to pH requirements.

2.4. FORMULAE

The following formulae are used in the calculation:

$$A = A_0 \exp[(-0.693 \times t)/T] \tag{1}$$

where

A is the activity after time t (cpm),
A_o is the initial activity before treatment at t_o (cpm),
t is the time taken (t-t_o) (sec), and
T is the half-life (sec).

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

where

A is the average reading for sample,
B is the average reading for background,
E is the efficiency of the detector (34% for Canberra Model 2400),
V is the sample volume (mL),and
Min is the counting time (minutes).

$$Decontamination\ factor\ (DF) = \frac{Activity\ of\ the\ effluent\ before\ treatment\ (counts/sec)}{Activity\ of\ the\ effluent\ after\ treatment\ (counts/sec)} \tag{3}$$

$$Percentage\ radionuclide\ removed = \frac{Original\ activity - Final\ activity}{Original\ activity}\times 100 \tag{4}$$

$$Distribution\ coefficient = \frac{(initial\ activity)-(final\ activity)}{(final\ activity)}\times\frac{total\ volume\ of\ the\ solution\ used\ (mL)}{mass\ of\ the\ soil\ used\ (g)} \tag{5}$$

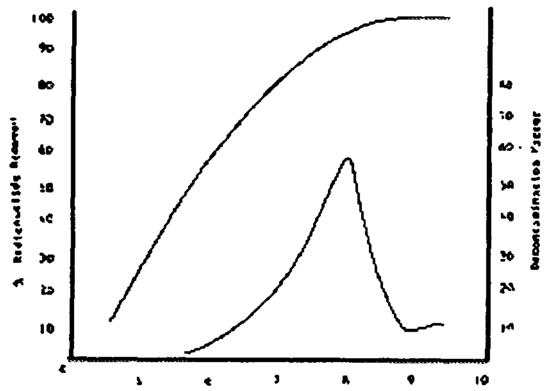

FIG. 1. % radionuclide removed against pH
Decontamination factor against pH

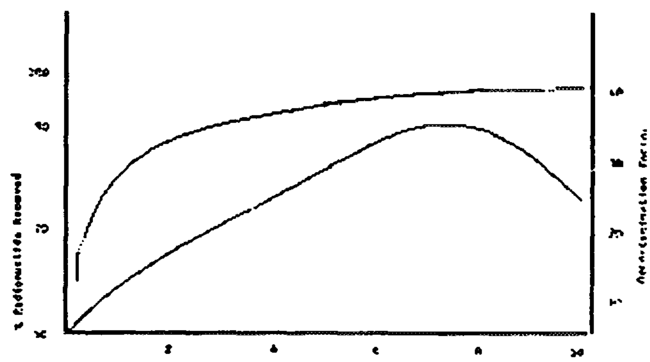

FIG. 2. % radionuclide removed and decontamination factor against alum (mL)

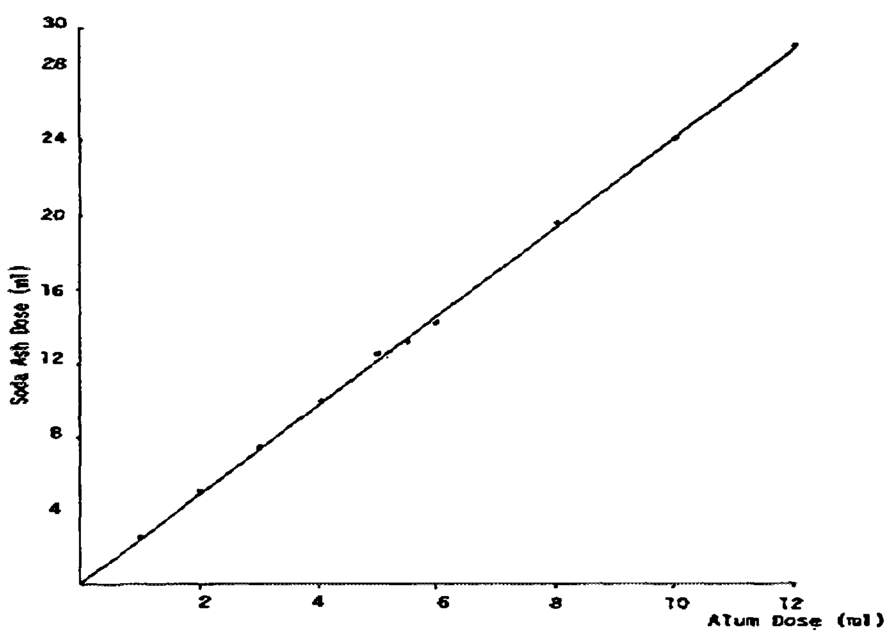

Fig. 3. Variations of alum and soda ash to maintain optimum pH8

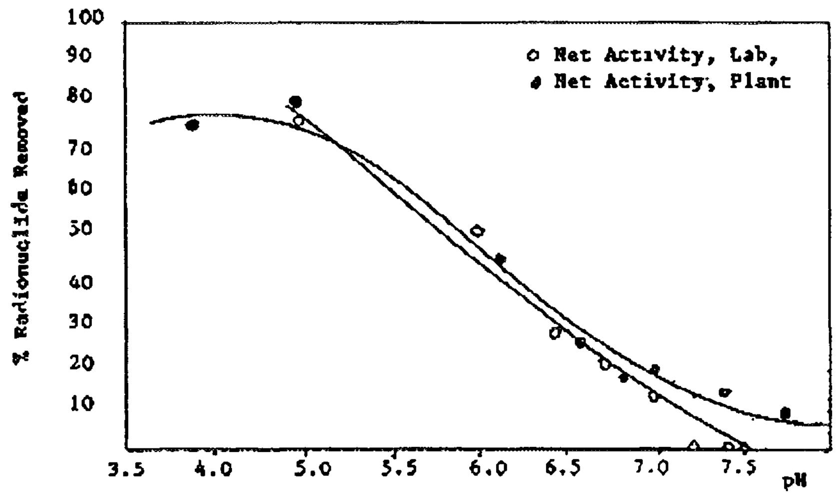

FIG. 4. Comparison of treatment in the laboratory and Low Waste Treatment Plant

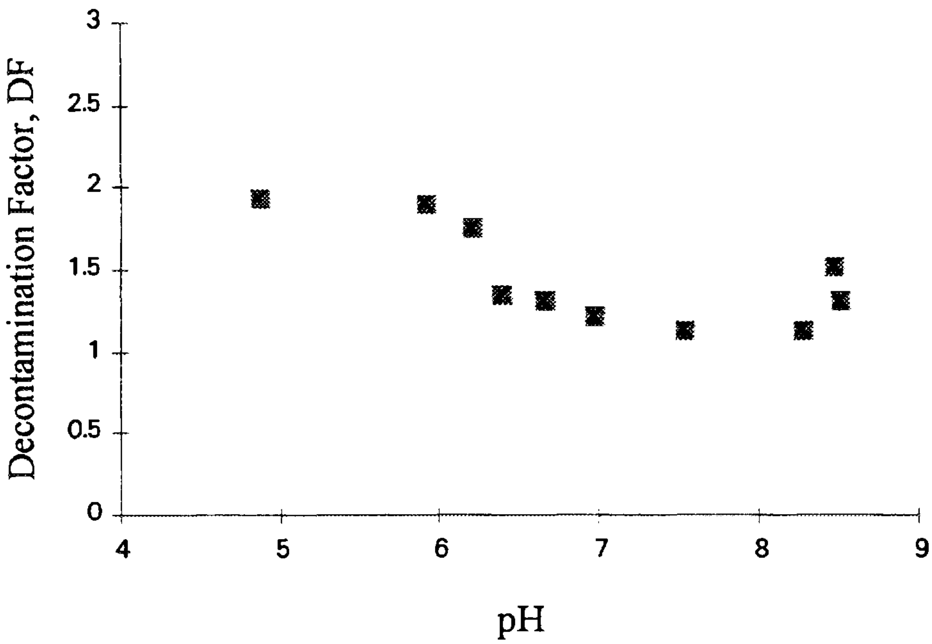

FIG. 5. Chemical treatment using alum as coagulant.

3. RESULTS AND DISCUSSIONS

Figure 1 shows that optimum pH value corresponds to the highest decontamination factor for removal of ^{65}Zn from the effluent. After pH8, the curve descends which shows there was a decrease in the percentage of the radionuclide removed. Experiments showed that alum and soda ash affected the size of particles produced. The particle size was reduced when reduced amounts of soda ash dosage were used. This produced lesser precipitation and caused smaller decontamination factor values. Increasing the amount of soda ash led to the increase of colloid particles produced.

Figure 2 shows the optimum alum dosage was 7 mL. The percentage of the radionuclide removed was nearly optimum at 95%. Major portions of the ^{65}Zn radionuclides were removed from the effluent. Figure 3 shows how the chemicals dosages of alum and soda ash were varied to maintain the optimum pH 8 condition. Figure 4 shows both plant and lab scales treatments for removal of ^{99}Mo. There are two curves

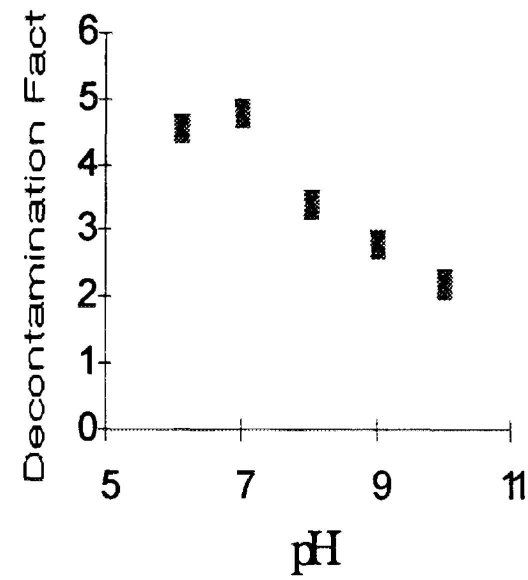

FIG. 6. Chemical treatment using ferric chloride as coagulant.

with little differences at different pH conditions. The jar test experiments at a laboratory scale were valid for the extrapolation to the plant scale treatment. Treatment of 1000 mL effluent on a laboratory scale, with similar conditions, methods and chemicals was scalable for the effluent treatment in the plant. The amount of the chemicals needed for the plant scale was calculated from the ratio obtained on the laboratory scale. The maximum pH range for removing about 80% of ^{99}Mo was between 4 and 4.5. Since the pH of the treated effluent was lower than the permissible discharge limit (i.e., pH6-9), pH adjustment was performed with the addition of soda ash.

Figure 5 shows the removal of ^{125}I using alum that produced the highest decontamination factor 2 with pH range 5.0 to 6.0. The value decreased between pH 6 to 8 and slightly increased again after pH8. Figure 6, between pH6 to pH7, the highest decontamination factor value determined was 5.0 using ferric chloride. The value decreased when the pH values increased.

4. CONCLUSION

From the experiments conducted, several conclusions can be made:

(1) The optimum pH for removing ^{65}Zn from the effluent is about 8;
(2) The optimum alum dosage for removing ^{65}Zn from the effluent is 7 mL;
(3) Extrapolation of the jar test results is scalable to the plant scale treatment;
(4) For the treatment of the effluent containing ^{99}Mo, the optimum pH is in the range of 4.0 to 4.5;
(5) For ^{125}I removal using alum, the highest decontamination factor value was 2 with pH range 5.0 to 6.0. However using ferric chloride, the highest decontamination factor value determined was 5.0, for pH between 6-7.

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