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Study of the Treatment of the Liquid Radioactive Waste Nong Son Uranium Ore Processing

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Abstract. Liquid waste from Nong Son uranium ore processing is treated with concentrated acid, agglomerated, leached, run through ion exchange and then treated with H_2O_2 to precipitate yellowcake. The liquid radioactive waste has a pH of 1.86 and a high content of radioactive elements, such as: [U] = 143.898 ppm and [Th] = 7.967 ppm. In addition, this waste contains many polluted chemical elements with high content, such as arsenic, mercury, aluminum, iron, zinc, magnesium, manganese and nickel. The application of the triditional method as one stage precipitation or precipitation in coordination with $BaCl_2$ is not effective. These methods generated a large amount of sludge with poor settling characteristics. The volume of final treated waste was large. This paper introduces the investigation of the treatment of this liquid radioactive waste by the method of two stage of precipitation in association with polyaluminicloride (PAC) and polymer. The impact of factors: pH, neutralizing agents, quantity of PAC and polymer to effect precipitation and improve the settling characteristics during processing was studied. The results showed that the processing of liquid radioactive waste treatment through two stages: first stage at pH = 3 and the second stage at pH = 8.5 with limited PAC and polymer (A 101) resulted in significant reduced volume of the treated waste. The discharged liquid satisfied the requirement of the National Technical Regulation on Industrial Waste Water (QCVN 24:2009).

Keywords: uranium processing, liquid radioactive waste, treatment.

1. Introduction

The radioactive liquid waste from Nong Son uranium ore processing have the mainly chemical contents and radioactivity as in Table 1 [1].

The contents of many metals in this radioactive liquid waste were higher over thousands times than the limited content of these metals in the National Technical Regulation on Industrial Waste Water (QCVN 24:2009). The application of generally technology for treatment of radioactive liquid waste from uranium ore processing to this waste [2-6]: one stage precipitation at pH = 8.5 – 9.0 with the co-precipitation by the addition of $BaCl_2$ showed that any amount of sludge was generated and the settling ability of this sludge was so bad (after 3 days of settling, the volume of sludge was still equal about 80% of the original volume). For the preparation the radioactive liquid waste sample to investigate

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the factors which impacted to the effect of treatment processing, the authors of the paper had carried out the first precipitation at pH = 3 to reject the large amount of iron from the liquid waste.

Table 1. The mainly chemical contents and radioactivity of radioactive liquid waste from Nong Son uranium ore processing

No.	Analyte	Content (ppm)
1	Al	4980.267
2	Fe	4101.435
3	Cu	6.203
4	Zn	833.722
5	As	15.858
6	Pb	0.545
7	Mg	947.549
8	P	238.700
9	Mn	2827.324
10	Ni	13.931
11	Th	7.967
12	U	143.898
13	Ra	0.004
14	Hg	0.013
15	Total radioactivity α (Bq/l)	38.9
16	Total radioactivity β (Bq/l)	261.1

The solid waste of iron hydroxide get from this stage can be deal with as the normal industrial waste or can be reused as by-product. The liquid from the filter after the first precipitation will be used as the sample for study the impact of pH value, the content of the precipitate (polyaluminiumcloride – PAC) and the content of polymer A101 to the effect of treatment processing, to the settling ability of the sludge. Finally, the authors had proposed the flow sheet of treatment of the liquid radioactive waste Nong Son uranium ore processing. In this flow sheet, the two stages precipitation processing with the use of PAC and polymer A101 had used. The application of this processing showed that the treated water satisfied the requirements of QCVN 24:2009 [7] and can be discharged to the environment.

2. Materials and methods

Object of research: The liquid radioactive waste from Nong Son uranium ore processing is treated with concentrated acid, agglomerated, leached, run through ion exchange and then treated with H_2O_2 to precipitate yellowcake.

Collection documents and data: documents and data for research were received from the documentation of the International Atomic Energy (IAEA) and the Vietnam Atomic Energy Institute; The reports, curriculums on the management of radioactive waste of The Institute for Technology of Radiaoactive and Rare Elements; The science books, newspapers, magazines related topics exploitation, collected through the Internet.

Analysis samples in the laboratory: Determination of uranium by the photometric method on Digital spectrophotometor 2000RS. Thorium and radium analysis on ICP - MS. The content of metal ions of treated water was control by ICP-MS and the total alpha activity, the total beta activity were measured by alpha/beta analyzer. The comparison between the analytic data with the limited content of factors in the National Technical Regulation on industrial waste water (QCVN 24:2009).

Experimental approaches: the study was the impact of pH value, the impact of PAC content and the determination of the suitable content of polymer A101 with the circulation time, the speed of the stir to the effect of the treatment processing. Propose the suitable factors for the treatment processing of radioactive liquid waste of Nong Son uranium ore processing.

3. Results and discussion

3.1. The impact of pH value to the effect of the treatment processing

In these experiments, the sample was the filtrate from the first precipitation at pH = 3. The primary turbidity of the sample was measured by HACH Spectrophotometer DR2010 and has the value of 1308 (Co – Pt). Using NaOH to correct the value of pH to 4.5, 5.0, 6.0, 7.0, 8.0, and 9.0, respectively; Stir the samples in 5 minutes with the speed of propeller of the mixer about 100 r/m. In these experiments, we keep the same PAC content (250 mg PAC/l) for every experiment. Continue the stir for 15 minutes with the speed of 20 r/m. Finish the experiments, keep the settling of the sludge for 30 minutes. Decant the treated water and measure the turbidity of its. The results were showed in the Table 2 and the Fig. 1.

Table 2. The impact of the pH value to the effect of the treatment processing

No. experiment	1	2	3	4	5	6
pH value	4.5	5.02	6.0	7.02	7.98	9.0
Turbidity (Co-Pt)	137	21	35	31	14	30
Effect (%)	89.5	98.4	97.3	97.6	98.9	97.7

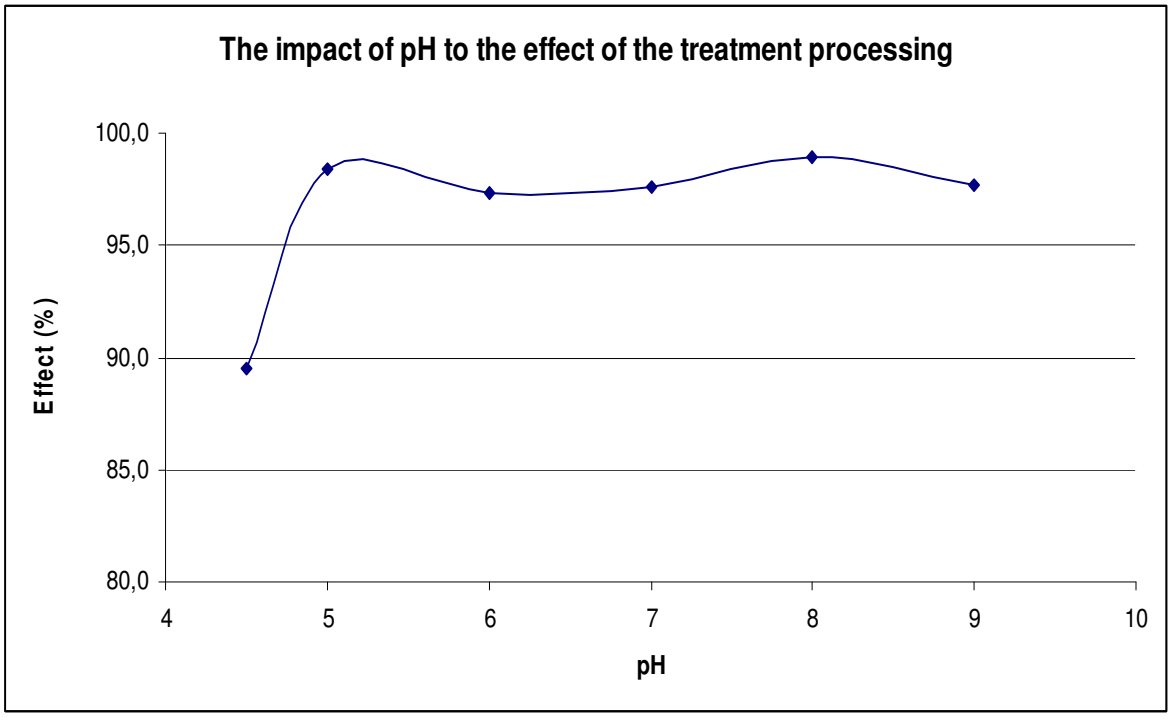

Fig 1. The impact of pH to the effect of the treatment processing.

Base on the results in the table 2 and the fig.1, we find that the pH value from 5 to 9 had a good effect to the treatment processing. The effect of the processing is about 97.3 – 98.9 %. So we chose the pH value of 8 for subsequent experiments.

3.2. Study the impact of PAC content to the effect of treatment processing

In the subsequent experiments, the pH value of each experiment was keep at the value of 8. The contents of PAC were changed to: 125, 250, and 375. 500, 625, 750 (mg/l) respectively the procedure of each experiment was carry out the same as in the above series. The results were showed in Table 3 and Fig. 2.

Table 3. The impact of PAC content to the effect of treatment processing

Sample	**1**	**2**	**3**	**4**	**5**	**6**
PAC content (mg/l)	125	250	375	500	625	750
pH value	7.95	7.98	8.02	7.96	7.98	9.0
Turbidity (Co-Pt)	32	20	35	17	32	42
Effect (%)	97.6	98.5	97.3	98.7	97.6	96.8

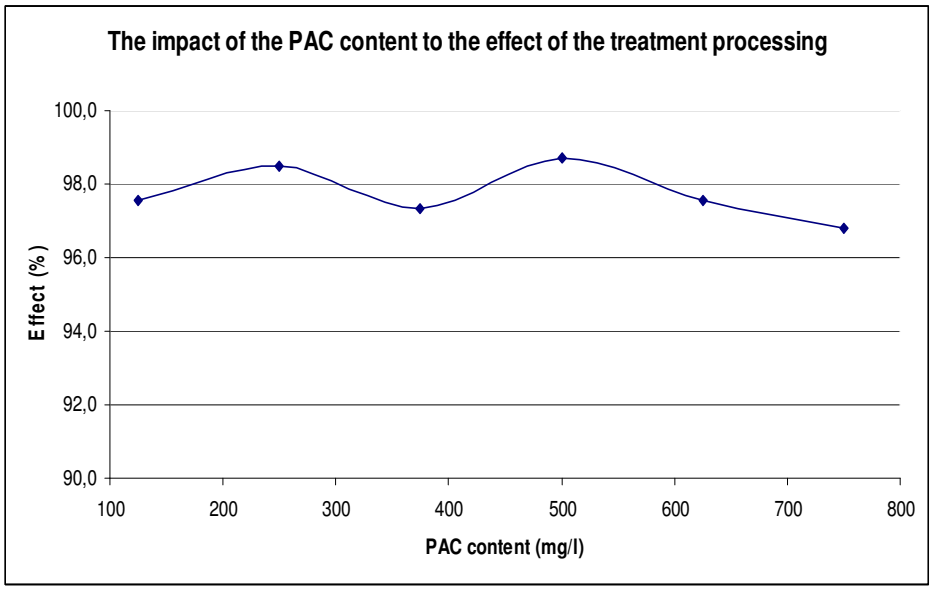

Fig. 2. The impact of PAC content to the effect of the treatment processing.

From the results in the Fig. 2 we recognized that which the PAC content from 125 – 800 mg/l, the effect of the treatment processing was from 97 – 99 %.

3.3. The determination of the suitable content of polymer A101

In the experiments of this part, we chose the pH value was 8 and the PAC content was 250 mg/l. The contents of polymer A101 were changed: 2.5, 3.75, 5.0, 6.25, 7.5, 8.75 (mg/l) respectively. The procedure of each experiment was the same as the before. To compare between these experiments we determine the time for settling the sludge from the primary volume to the 1/4 its volume. The results were showed in Table 4 and Fig. 3.

Table 4. The impact of the content of polymer A101 to the settling time of the sludge

Sample	1	2	3	4	5	6
Polymer content (mg/l)	2.5	3.75	5.0	6.25	7.5	8.75
pH value	7.96	7.99	7.98	7.96	7.94	7.96
Settling time (m)	9	6.5	5.2	5.0	5.5	5.0

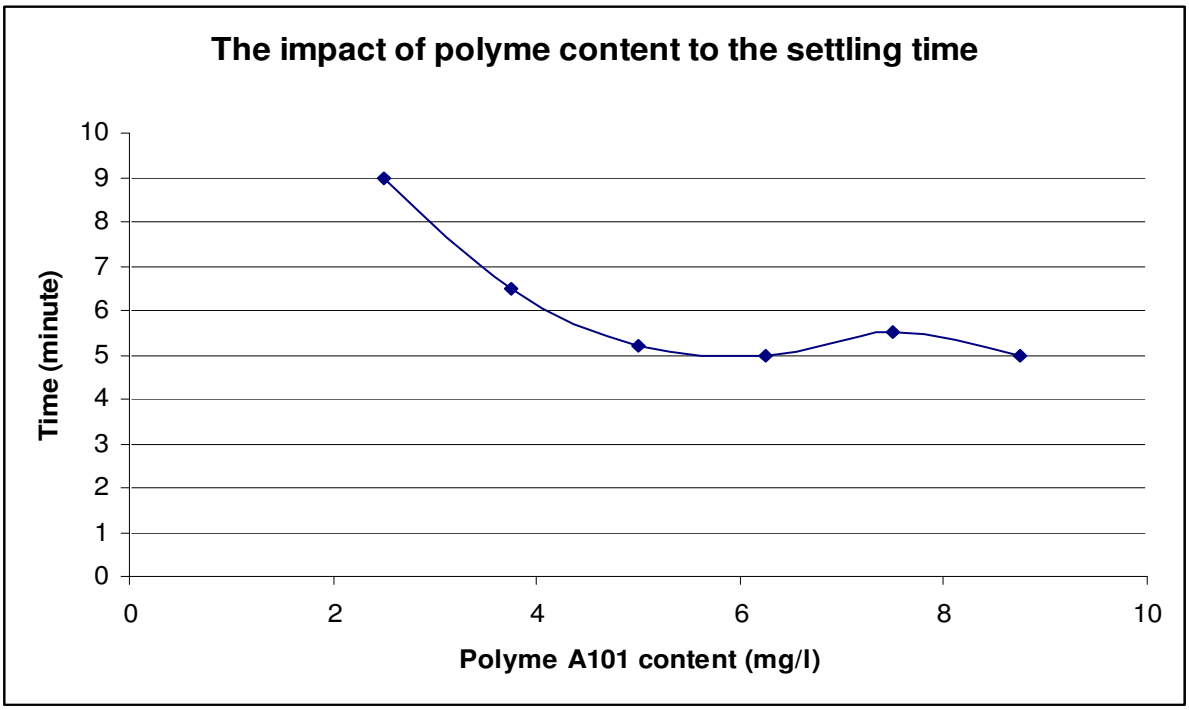

Fig. 3. The impact of polymer content to the settling time of the sludge.

Base on Fig.3 we find that when the content of the polymer > 3.75 mg/l the settling time changed not significant and had the value of 5 – 6 minutes. In the technological condition, this time can be acceptable. So we can choose the content of the polymer A101 was 5 mg/l.

3.4. The flow sheet of the treatment of the liquid radioactive waste Nong Son uranium ore processing

The flow sheet of the treatment of the liquid radioactive waste Nong Son uranium ore processing was on Fig. 4.

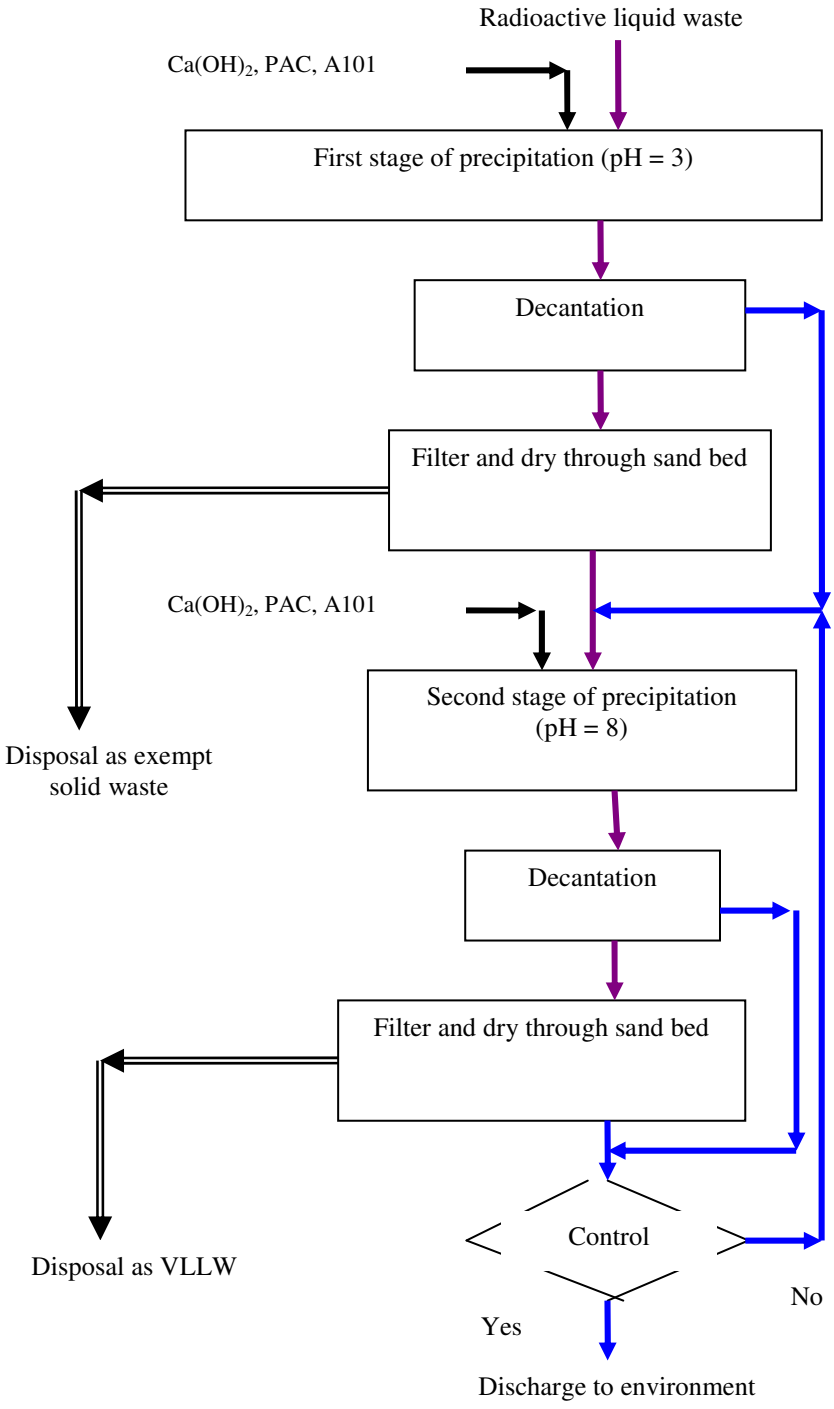

Fig. 4. Flow sheet of treatment of the liquid radioactive waste Nong Son uranium ore processing.

After studying the impact of pH value, content of PAC, content of polymer, we had proposed the suitable factors for the treatment processing of radioactive liquid waste of Nong Son uranium ore processing as following:

- The flow sheet with two stages of precipitation, at pH = 3 and at pH = 8;
- The PAC content: 250 mg/l;
- The polymer content: A101: 5 mg/l;
- The circulation time in the area was about 5 minutes with the speed of the stir about 100 r/m (for precipitation) and was about 15 - 20 minutes with the speed of the stir about 20 r/m (for agglomeration).

After the application of the flow sheet, the content of metal ions of treated water was control by ICP-MS and the total alpha activity, the total beta activity were measured by alpha/beta analyzer. The results of analysis were showed in Table 5.

Table 5. The comparison of the factors in primary waste water, treated water and QCVN 24:2009

Analyte	The content (mg/l)		QCVN 24:2009 (type B)* (mg/l)
	Primary waste water	Treated water	
Al	4980,267	0.087	-
Cr (III)	12.144	0.001	1
P	238.700	0.334	6
Cd	1.394	< 0.0001	0.01
Fe	4101.435	0.683	5
Ni	13.912	< 0.0001	0.5
Cu	6.203	0.128	2
Zn	833.722	0.014	3
As	15.858	0.002	0.1
Pb	0.545	< 0.0001	0.5
Mn	2827.324	0.009	1
Mg	947.549	9.904	-
Th	7.967	< 00001	-
U	143.898	0.380	-
Total radioactivity α (Bq/l)	38.92	0.03	0.1
Total radioactivity β (Bq/l)	261.16	0.94	1.0

** Type B: the value of content of contaminated analyses in the industrial waste water were allowed to discharge to the receipted resource, which will be not use for purpose of life water provision.*

The comparison between the analytic data with the limited content of factors in the National Technical Regulation on industrial waste water (QCVN 24:2009) showed that the treated water fully satisfied this National Technical Regulation.

4. Conclusions

The flow sheet for treatment radioactive liquid waste from uranium ore processing had been proposed in this paper had a good result to reduce the volume of the sludge, the solid waste get from the first stage of precipitation have the total radioactivity about 1 Bg/g and can be disposed as the normal industrial waste or reused as a by-product. The sludge at the second stage of the precipitation had a good settling ability and easy to decant and to filter. The final effluent from this processing is satisfied requirements of National Technical Regulation on industrial waste water (QCVN 24:2009).

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