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Study on Developing Safety Infrastructure for Mineral Processing Waste (NORM Waste) and Contamination Monitoring at the TINT Rare Earth Research & Development Center, Khlong 5, Pathumthani, Thailand

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Abstract. The objective of this study is to prepare all technical and administrative actions leading to the release of the disused radiological facilities from regulatory control and safe management of radioactive waste. In the first phase, the study covers: the preliminary site characterization, waste characterization, the preparation for authorization of the foreseen strategy and activities. The first phase study shows that several areas of the TINT's Rare Earth Research & Development Center are contaminated, such as the U/Th extraction, monazite processing and NORM residues storage.

1. Introduction

What is NORM? NORM stands for "naturally occurring radioactive material" or in other words, a substance that naturally contains one or more radioactive isotopes, also called radionuclides. NORM waste typically is produced by an industrial, mining, or manufacturing process that uses NORM.

In in this paper, the NORM wastes were generated from the monazite processing and the U-Th extraction pilot plans from the Rare-Earth Research and Development Center (RE R&D Center), Thailand Institute of Nuclear Technology (TINT). In Thailand, Monazite (Ce, La, Nd, Y, Th)PO4, occurs in heavy-mineral sand deposits, vein type deposits in low grade tin ores from the south of Thailand, with a rare earth content of about 60.20 % REO [1]. Rare earth mineral concentrates are chemically processed to extract intermediate groups of mixed rare earth compounds. In general, chemical treatment of mineral concentrates derived from hard rock deposits may start with roasting in air (calcining) to drive off carbon dioxide and oxidize cerium to the tetravalent state. This is, in many steps, grinding, caustic leach then followed by treatment with hydrochloric acid to dissolve non-cerium rare earths, yielding a marketable cerium concentrate which can be used directly as a low value product or further separated into high purity individual rare earths. Figure 1 shows the monazite processing chart from IAEA Safety Series [1].

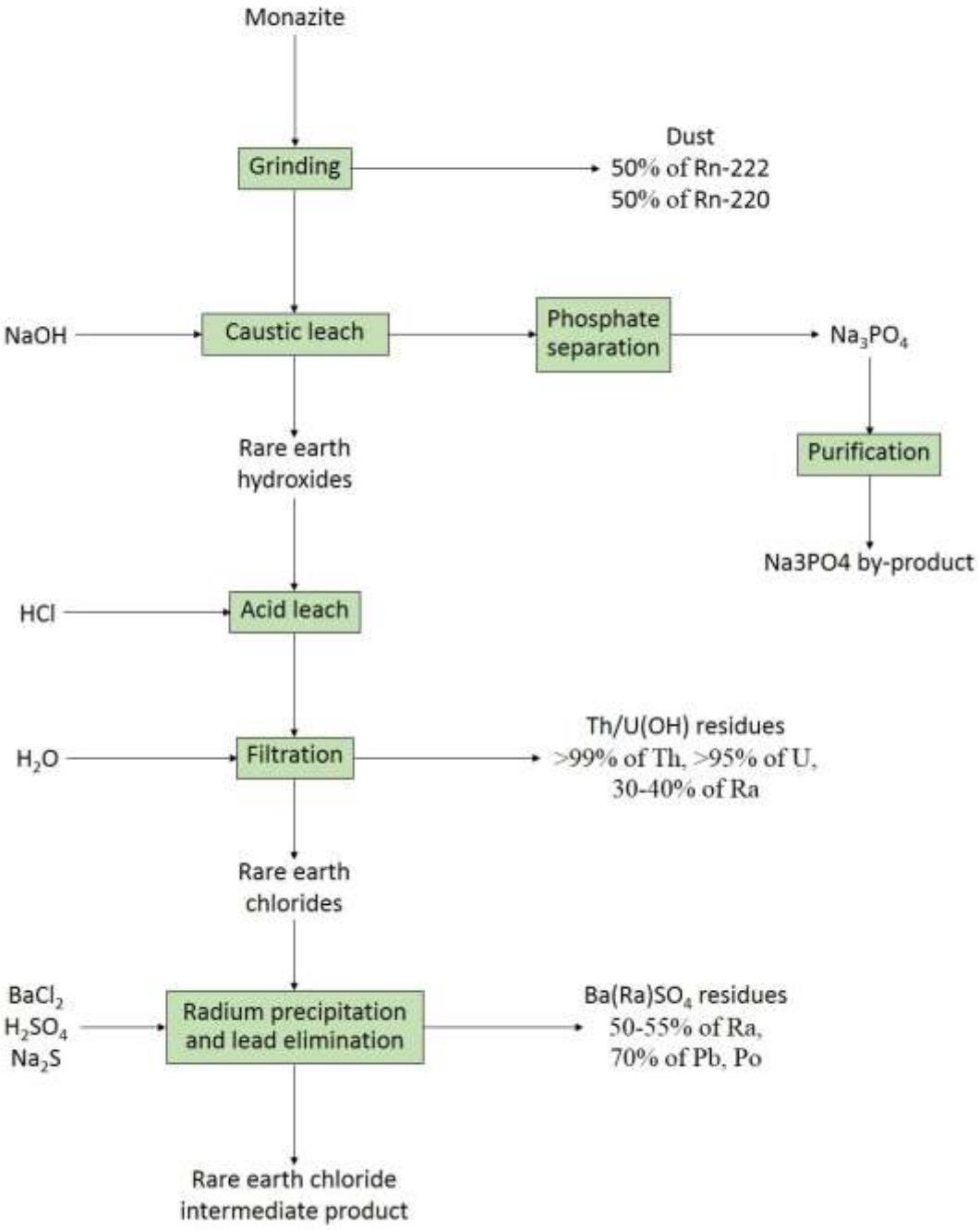

Figure 1. Monazite processing chart [1]

2. Historical Information and Infrastructure of the RE R&D Center

The RE R&D Center is located at Khlong Luang district, Pathumthani province, Thailand, about 40 km away from the TINT, Bangkok. This center was operated by Office of Atom for Peace during 1995 – 2005 for extraction of rare earth elements from monazite sand and also the U-Th extraction activities. After the re-organization on December 2006, the RE R&D Center has been transferred to the TINT. However, the rare-earth and monazite processing and the U/Th extraction plants have been stopped in operation since 2005. Later, in 2011, the heavy flood occurred in Thailand, the pilot plants and area around were unfortunately flooded. It was possible for the contamination with naturally occurring radioactive materials (NORM).

There were about 18 buildings for several activities. The building No. 8 consisted of the monazite processing and uranium and thorium processing plants which were the main building for radiological activities. The building No. 9 was the rare earth processing plant for extraction of rare earth elements. The building No. 14 was the storage facility for uranium and thorium cake. The building No. 18 was monazite storage which contains a huge quantity of monazite sand. The figure 2 shows the map of Rare Earth Research Development Center. Up to now, those building filled with the same instruments and materials as before, safety measures are not in place. There is no classified waste. There are mixed-items, such as instruments, chemicals, NORM residues and other infected waste from birds accumulated in those facilities.

Currently, there are a lot of NORM residues in drums onsite. The radiation safety infrastructure for NORM residue management is recommended to be seriously concerned. So far, the radioactive waste, both solid and liquid wastes are awaiting to be managed in proper ways.

In addition, the historical activity concentration of radon in each building was collected during this study as shown in Table 1 and Table 2.

Figure 2. Map of Rare Earth Research Development Center

Table 1. Indoor Rn-222 and Rn-220 concentration at the Rare-Earth Research and Development Center (measured date: Feb 2010) [2]

Location	Range of concentration ($Bq.m^{-3}$)	
	Rn-222	Rn-220
Rare Earth Processing Building 9	3 – 20	10 – 30
Monazite/ U-Th Processing Building 8	20 – 70	490 – 2200
U-Th Cake storage Building 14	160 – 442	870 – 5150

Remark: US EPA action level of radon in building should not exceed 4pCi/l, 148 $Bq.m^{-3}$ [3]

Table 2. Concentration of radon in workplaces at the Rare-Earth Research and Development Center (measure dated: March 2013) [4]

Location	Rn concentration in the air ($Bq.m^{-3}$)	Temp. (^{O}C) (RAD7)	RH (%) (RAD7)
1. Building No. 8	180 ± 10	31.6	12
2. Outside of Building No.14	52 ± 7	35.6	11
3. inside of Building No. 10	16 ± 4	35.3	11

Remark: Build No.10 is located in the opposite side of No.14 (this building has no activity)

3. Scope of Work in Phase 1

The work in phase 1 covers on study the TINT Rare Earth Research & Development Center's current safety situation. The scope of work in phase 1 is as followings;

- Onsite Survey: dose rate, and dose mapping
- Checking contamination at the monazite/ U-Th processing building (Building 8)
- Searching for NORM residue onsite
- Characterization of NORM residue

4. Method

4.1. Survey on Dose rate and Contamination Monitoring at the facilities

- The dose rate survey meter and the contamination monitor (which was able to detect alpha radiation) were used for survey the area in building 8, 9 and 18 (monazite storage building).
- The dose rate survey meter, Ludlum model 14C was used for measuring the exposure dose
- The contamination monitoring at the building No. 8, was conducted by direct method. The contamination survey meter, Ludlum model 12 with probe model 44-9, were used for measuring contamination.

Direct method is the measurement of radiological contamination by using the detector measure on the area directly. The distance from the detector from the target area is about 0.5 cm for alpha and 2.5 cm. for beta/gamma.

The area in the building No.8 was divided into 2 sections, *i.e.* the uranium and thorium processing and monazite processing facilities. The dose and contamination mapping method was performed for both facilities. The grid-method was applied coordinate to measure the highest contaminations in each grid. In the same time, the number of drums which contain liquid, sludge and residues were also counted.

4.2. Monitoring environmental water sample onsite

In order to monitor environmental water sample onsite, six water samples were collected randomly from different ponds as shown in figure 3. Water was collected from ponds and kept into 5 liters plastic bottle using manual procedure. The samples were transferred to the Radioactive Waste Management (RWM) Center's laboratory for analysis.


Pond no.1


Environmental monitoring point 1


Pong for sediment suspensions


Pond no.7


Channel beside the pond 7

Figure 3. Sampling water samples from ponds onsite

4.2.1. Gross Alpha Beta Analysis. The gross alpha and gross beta activity concentration measurement was carried out by gas flow proportional counter, using Low-level Gas Proportional Counter Berthold LB770. All samples were prepared by EPA method (EPA method 900.0). The counting time was 40 minutes for each counting period.

4.2.2. Gamma Analysis. 1 liter of each sample was poured into a Marinelli beaker in preparation of gamma spectrometry analysis. After properly tightening the threatened lid, a Marinelli beaker were sealed with adhesive tape and left for at least 4 weeks (>7 half-lives of ^{222}Rn and ^{224}Rn) before counting by gamma spectrometry in order to ensure that the daughter products of ^{226}Ra up to ^{210}Pb and of ^{228}Th up to ^{208}Pb achieve equilibrium with their respective parent radionuclides. The mass activity concentration of ^{40}K, ^{226}Ra and ^{232}Th determined gamma-ray spectrometry, using an ORTEC Hyper Purity Ge (HPGe) detector of 60% relative efficiency, coupled to a PC based digital analyser system employing ORTEC Gamma Vision software. Count time was in excess of 80,000 sec per sample, providing results with analytical precision of around 1% at the 95% level of confidence.

4.3. Characterization of solid waste (NORM residue)
During the survey of the site, NORM residue samples were collects from several places (7 points around treatment ponds and monazite storage). Solid samples were analyzed by using XRF technique provided by Physics and Engineering Group, Nuclear R&D Division, TINT. The XRF technique was applied for 60 seconds in TestAll Geo mode for each sample to find the elemental composition.

4.4. Characterization of liquid waste in drums stored at building No. 8
- Collecting samples of liquid waste from drums at the building No 8
- Analysis by Gamma Spectrometry (the same methods as in 3.3)

5. Results and Discussions

5.1. Survey on Dose rate and Contamination Monitoring at facilities

5.1.1. Dose rate and contamination map at the building No. 9. The dose rate of the over-all area in building 9 where the rare earth processing plant used to be operated, was found in the level of background (1 μSv/hr), and no contamination.

5.1.2. Dose rate and contamination map at the building No. 18. The maximum dose rate of the wall of building 18 was found to be very high dose rate, 62.2 μSv/hr. The average of dose rate around the wall was about 30-60 μSv/hr. It is about 30-60 times of background level (1 μSv/hr) at the RE R&D Center. Figure 3 shows that the building no.18 is full with monazite sand, and our radiation safety officer did not allow us to get inside due to the hazard of radon gas because the room was closed for a long time.

Figure 4. Dose rate of the wall of building 18

5.1.3. Dose rate and contamination map at the building No. 8. Building No. 8 was separated to 2 sections: the uranium and thorium processing section and the monazite processing section. The grid is applied to each section to estimate dose rate and contamination, including the volume of materials (in 80 Liter drum) in each square. The results of dose rate and contamination are shown in Figure 5 and Figure 6, respectively. Wide areas of contamination cause high dose rates in the building No. 8, mainly in the uranium and thorium processing section. The highest contamination value is at the A1 (100,000cpm in the map).

In addition, there are about 185 containers which contain NORM residues and chemical solvents in the U/Th processing section.

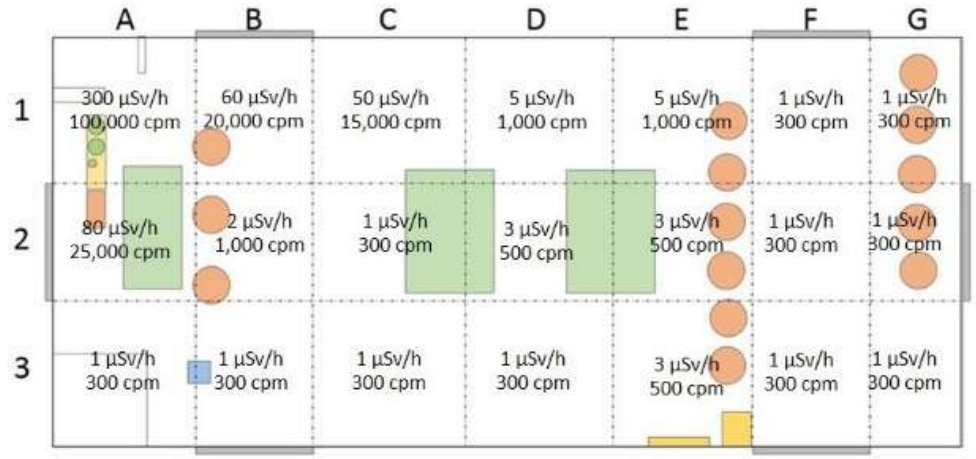

Figure 5. Dose rate and contamination map of uranium processing

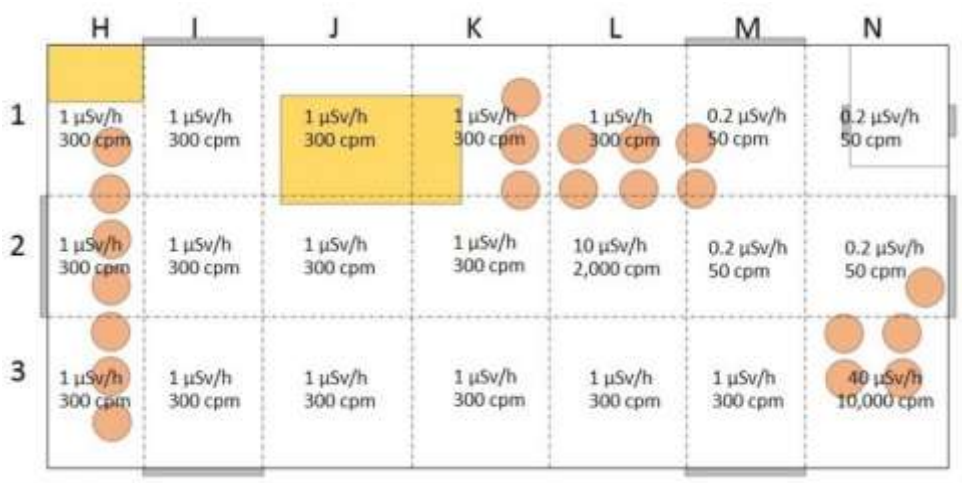

Figure 6. Dose rate and contamination map of monazite processing

5.2. Monitoring environmental sample onsite

Gross alpha- gross beta of water samples from each pond were analyzed, the results were shown in Table 3.

Table 3. Gross alpha-gross beta of water samples from ponds onsite

No	Sample	Activity Concentration (Bq/L)	
		Gross Alpha	Gross Beta
E1	Pond 7	< 0.018	0.715
E2	Channel beside the pond 7	< 0.018	0.878
E3	Pond (for sediment suspensions and filtration)	< 0.018	0.841
E4	Environmental monitoring point 1	< 0.018	0.426
E5	Pond 1	< 0.018	0.922
E6	Environmental monitoring point 2	< 0.018	0.508
Baseline Surface water before operation (1991) [5]		0.037	0.274
Detection limit, DL (Bq/L) (as July 2016)		0.018	0.012

Also the samples had been left for 4 weeks and were analyzed by using HPGe spectrometer for 24 hours to find the concentration of Ra-226, Th-232 and K-40 as shown in Table 4.

Table 4. Radionuclides and activity concentration in surface water from the ponds onsite

No	Sample	Activity Concentration (Bq/L)		
		Ra-226	Th-232	K-40
E1	Pond 7	10.41 ± 2.84	1.66 ± 0.64	65.19 ± 5.31
E2	Channel beside the pond 7	4.15 ± 0.33	1.72 ± 1.22	66.69 ± 3.12
E3	Pond filter sediment suspensions	15.49 ± 2.95	1.15 ± 0.46	66.11 ± 3.23
E4	Environmental monitoring point 1	8.84 ± 2.39	1.63 ± 0.64	66.92 ± 3.12
E5	Pond 1	7.87 ± 2.34	2.36 ± 0.67	63.91 ± 3.12
E6	Environmental monitoring point 2	8.94 ± 2.34	1.28 ± 0.67	63.92 ± 3.12
Baseline Surface water before operation (1991) [5]		0.00126		

5.3. Characterization of solid waste (NORM residue)

The NORM residue samples were collected from several points in the area of the RE R&D center, mainly around the monazite storage and the treatment ponds. The XRF technique was applied. The results are shown in Table 5. The results show that the quantities of uranium and thorium were found in high value in the sample no 5, the sludge from P5 which was the evaporation pond for treatment of waste water from uranium and thorium processing section.

Table 5. Uranium and thorium containing in NORM residue samples

No	Origin of Sample	ppm	
		Th	U
1	Sludge from Pond 7	ND	18.92
2	Sludge from the side channel of Pond 7	11.39	14.21
3	Sand in the drum at the monazite storage	ND	ND
5	Sample from other R&E laboratories	51.27	ND
6	Sludge from Pond 5	1524.12	1981.78
7	Sample from other R&E laboratories	59.92	ND
8	CeO_2 residues	ND	ND

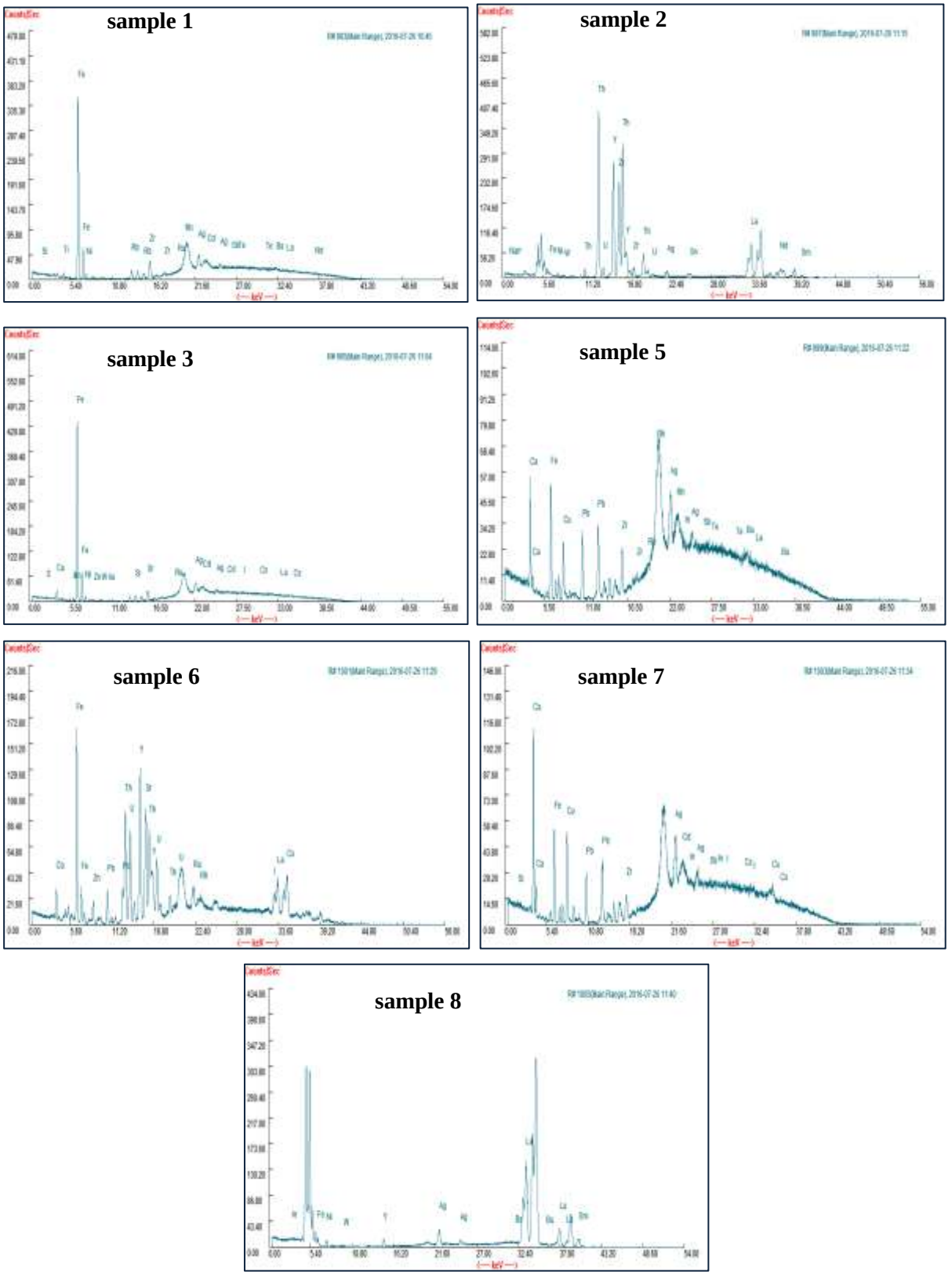

Figure 7. The XRF results from NORM residue samples

Note: The Y-axis is cps and the X-axis is energy of X-rays in keV

5.4. Characterization of liquid waste in drums stored at Building 8

Activity concentrations of liquid waste samples are as shown in Table 6. These results show that the high concentration of Ra-226, Th-232 and K-40 in liquid waste samples were found in most samples, they could be the contaminated solvents from the monazite processing. The different composition of these elements in contaminated waste samples was depend on monazite sand U-Th processing procedures.

Table 6. Activity concentration of liquid waste in drums stored at Building 8

Container no.	pH	Activity Concentration (Bq/l)		
		Ra-226	Th-232	K-40
1	2	1441.11 ± 114.72	19612.12 ± 42.5	1600.9 ± 49.5
3	5	8.87 ± 0.17	31.21 ± 1.84	66.81 ± 5.88
23	13	7.77 ± 3.35	4.15 ± 0.92	66.64 ± 3.81
24	1	1668 ± 104.51	16502.54 ± 47.82	1339.56 ± 54
25	1	683.76 ± 80.14	11373.05 ± 36.89	980.62 ± 41.19
48	1	-	32646.78 ± 51.91	-
51	1	653.91 ± 13.81	19584.13 ± 42.29	1558.32 ± 59.88
52	5	16 ± 8.53	65.87 ± 1.27	79.96 ± 5.54

6. Advices on safety infrastructures

Worker awareness and training are particularly important for supporting the introduction of radiation safety rules and for creating an understanding of the precautions embodied in such rules. Individual employee work practices may exacerbate dust generation**.** The general standard of housekeeping and spillage control also needs to be kept under regular review. Even when low activity concentration materials are handled, a reasonable standard of housekeeping may be necessary to ensure that dust and dirty from bird sham resuspension are adequately controlled. Very high standards would generally be required in the processing areas and storage areas where highly active materials, such as monazite sand and U cake/Th cake are stored.

7. Conclusion

This study is aimed to survey the storage area of NORM residue/ waste. We found contamination in some water samples from ponds and high contamination in liquid waste stored at Building8. The regulatory controls of NORM should be strengthened, e.g. management of NORM residues, residues disposal, environment monitoring, and effluent (processing water, gas) discharge to environment. There should be the clear declaration and segregation of radioactive and non-radioactive waste. The comprehensive plan should be draw up to balance of prevent contamination with workers. The survey of the whole site should be conducted. The contamination zone found should be clean up or remediated.

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