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RESEARCH ARTICLE

Assessment of the radiation exposures during separation of rare earth elements from monazite mineral

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Abstract

Through the separation of rare earths elements REEs from monazite mineral, radiation exposures occur. This study estimated the values of these radiation exposures. The study was occurred through two different treatments of monazite, 10kg of monazite and 10kg of monazite with adding BaCl at the digestion stage. The activity concentration of radon gas varied between 0.81 and 14.4 (Bq/m³) with an average of 8.72 (Bq/m³) while the activity concentration of thoron gas varied between 56.88 and 114.7 (Bq/m³) with an average of 76.14 (Bq/m³). Uranium concentration values in the air of the various areas changed from 0.29 to 0.88 (Bq/m³) with an average of 0.39 (Bq/m³). The gamma equivalent dose rates varied from 0.11 to 0.21 (μSv/h) with average 0.15 (μSv/h). The total annual effective dose received by the workers at the the REES project has values from 0.46 to 12.46 (mSv) with average values of 3.21 (mSv).

It was concluded that the values of the occupational effective doses are within the dose limits at the different stages of chemical processing of monazite. However, it is recommended to make technical enhancement to reduce the U-content in the air of the REEs hanger.

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Introduction:-

Monazite mineral is an important resource of the rare earth elements REEs. These elements are known to have higher potential to be used in several industries [1]. During the separation of the REEs from monazite mineral, radiation exposures arise to the occupational workers due to the existence of the radioactive elements uranium, thorium and their decay chains.

Pillai [2] studied the presence of NORM in the rare earth minerals in varying concentrations is quite often significant enough to result in occupational and environmental radiation exposures during their mining, milling and chemical processing for the extraction of the rare earth elements and compounds. Depending on the monazite concentration in the raw sand, radiation exposures of the order of 0.13–1.00 man.mSv per tonne are involved in the mining and separation of monazite. The chemical extraction of rare earths from monazite involves occupational radiation exposures in the range 0.30–1.00 (mean: 0.64 ± 0.19) man.mSv per ton of rare earths concentrate.

The long-term health effects of exposure to thorium are of interest because of the possible increased use of thorium as an energy source in reactors using ²³²Th to produce ²³³U. Mortality is described in a cohort of 3039 men who were employed between 1940 and 1973 at a company involved in the production of thorium and rare earth chemicals from monazite sand. Based on deaths ascertained by the Social Security Administration and mortality rates for U.S. white males, the standardized mortality ratio (SMR) for all causes was 1.05 with 95% confidence

limits (95% CL) of 0.96 and 1.15. Much of the excess mortality was attributable to non-occupational motor vehicle accidents (SMR=1.64; 95% CL=1.16 and 2.23), but SMRs were also high for lung cancer (1.44; 95% CL= 0.98 and 2.02), pancreatic cancer (2.01; 95% CL=0.92 and 3.82), and diseases of the respiratory system (1.31; 95% CL=0.92 and 1.83). In a subgroup of 592 men who worked for at least one year in selected jobs (indicative of highest exposure to thorium and thoron) that was followed up more intensively, the SMR for pancreatic cancer was significantly elevated (i.e. 4.13; 95% confidence limits=1.34 and 9.63). The SMR for lung cancer was 1.68 (95% CL=0.81 and 3.09), while that for respiratory diseases was 1.20 (95% CL=0.52 and 2.37). Information on smoking habits in a sample of survivors suggested that smoking could have explained at least part of the excess mortality from lung and pancreatic cancer and from diseases of the respiratory system. Continued follow-up of the cohort through morbidity and mortality studies is needed to evaluate further the possible long-term effects of exposure to radioactivity and chemicals in the thorium extraction process [3].

One of the chief problems in industrial radiological health in Brazil concerns 420 people in a plant processing monazite sand, a rare earth mineral containing up to 6 per cent ThO₂ and 0.3 per cent U₃O₈. Costa-Ribeiro evaluated the Radiation hazards associated with ore concentration and chemical treatment were assessed by sampling and measurement of airborne radioactivity, ²¹⁰Po bioassay in urine and chromosomal analysis in peripheral blood lymphocytes collected from plant personnel. Results showed that ²¹²Pb (t_{1/2} = 10.64 hr) airborne concentrations range from 0.002 up to 3.9 pCi/l. A size distribution analysis showed that in 88 per cent of ²¹²Pb-bearing aerosols the mean count diameter is below 0.4 [μm]. Calculated values of airborne long-lived activity as ²³²Th ranged from 6 x 10⁻⁶ up to 1 x 10⁻⁴ pCi/l. A few workers had a slight increase in the urinary concentration of ²¹⁰Po, although results did not show a statistically significant difference between these values and those found in control population. Chromosomal analysis revealed a statistically significant difference in the yield of aberrations for the ore mill workers in one of the plant sections, showing the highest airborne ²¹²Pb concentration [4].

In India, rare-earth compounds are produced from the beach sand mineral monazite. Caustic digestion of the mineral followed by selective acid extraction is the method used to separate composite rare-earth fraction. The composite rare-earth chloride contains low levels of natural radionuclides and is the starting material for individual rare-earth compounds which have wide applications. Activity concentrations in composite rare-earth compounds such as chlorides, fluorides, carbonates and oxides of Ce, Nd, Pr, Sm, Gd, etc. are presented in the study. The external gamma exposure rates and airborne activity due to thorium and thoron progeny in the occupational environment are studied. The activity levels in liquid effluent are presented. The potential individual occupational dose is estimated to be 1.9 mSv per annum [5].

Chemical processing of monazite:-

Rare earths have been obtained from monazite concentrate through digestion with 98% sulphuric acid at 200–220°C. The mineral is decomposed exothermically, forming a pasty mixture of sulphates and acid sulphates suspended in phosphoric acid and excess sulphuric acid. The rare earth chloride solution contains small amounts of impurities such as thorium, uranium, lead, iron and radium.. These are removed through a ‘deactivation and lead elimination’ process. Barium chloride and sodium sulphate or sulphuric acid are added to co-precipitate lead and other heavy metals along with the barium–radium sulphate formed. Later, sodium sulphide is added in excess to precipitate lead as lead sulphide as well as thorium, uranium and iron. The combined precipitates are removed by filtration as a barium sulphate cake containing radium and lead. This residue, which is sometimes referred to as ‘mixed cake’ or ‘mesothorium cake’ is reported in one instance to contain 1.07% thorium, 0.076% uranium, 27% barium sulphate and 21% rare earth phosphate [6,7]. Modifications to the deactivation and lead elimination process, involving multiple deactivation steps and pH control, have resulted in lower radioactivity levels in the final rare earth chloride product. Multiple deactivation involves further additions of barium chloride and sodium sulphate, resulting in a larger volume of material and, consequently, lower concentrations of radionuclides in the residue as well as in the product [1]. The flow sheet describes the different stages of the process is shown in Fig (1).

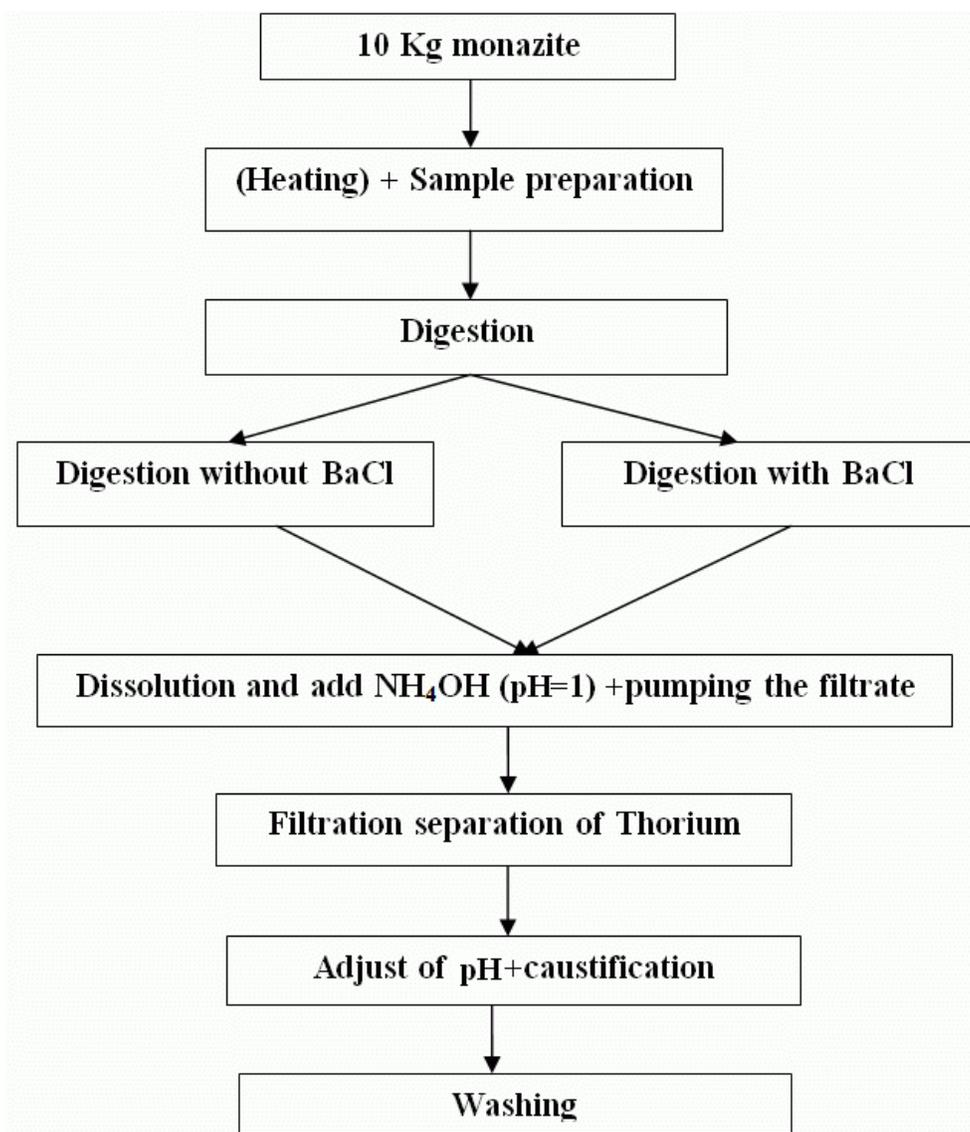


Fig (1): Flow sheet of monazite describes the different stages of the process.

This paper discusses comprehensively the different processes adopted in chemical separation of rare earths from monazite. Estimation of the radiation hazards that occupational workers are exposed to through this processing will be carried out. An approach to minimize the radiation exposures through the chemical extraction of rare earths from monazite will be discussed.

Experimental methods and techniques:-

The measurements to estimate radiation effective doses were carried out at the hanger of the rare earth elements project (REEs) at the Nuclear Materials Authority (NMA), Egypt. The source of monazite mineral is the beach deposits at Abu Khashaba, northern Nile coast, Egypt. Monazite mineral was concentrated from the beach deposits at the project of concentration and separation of black sands, Egypt.

Measurement of equivalent dose (H):-

Measurement of external exposure was estimated by measuring gamma rays using a portable survey meter. Measurements were carried out through the processing of monazite by using survey meter Alnor RDS-100. Direct reading of the equivalent dose rate (H) ($\mu\text{Sv/h}$) is recorded.

Estimation of internal exposures:-

Internal exposure occurs as a result of the inhalation of radon and thoron gases in the air and due to the uranium in the dust particles.

- **Measurements of radon and thoron concentrations**

A SARAD, RTM-1688 device was used to measure the radon and thoron gases concentrations C_{Rn} and C_{Th} . The device collects an air sample via an internal pump inside its chamber. Concentrations of these gases are displayed on the screen after a desired time after counting the alpha particles emitted from the studied gases and their decay products in the chamber.

- **Measurement of the Uranium concentration in the air**

Concentrations of uranium in the air dust are measured by collecting an air sample by a sampler on a high efficiency filters for 10 minutes. The α -particles will be counted after at least three days so that the short-lived radon and thoron decay products are completely decayed. Uranium content in the air C_U is calculated by the equation [8]:

$$C_U = 2.2 \times 10^{-7} (R-B) (1+S) / E t v \quad (1)$$

C_U : uranium concentration in ($\mu\text{Ci}/\text{cm}^3$),

R: α - count rate in (cpm),

E : counting efficiency,

v : volumetric sampling rate in (l/min),

t : sampling time,

B : background (cpm),

S : is the self absorption of the filter = 0.09.

Results are converted into (Bq/m^3).

Results and discussion:-

During the chemical processing of monazite to extract the REEs there are radiation exposures both externally (γ -rays) and internally; radon, thoron gases and U-content in the air of working area.

External exposure from gamma rays:-

Effective dose rate is calculated depending on the configuration of exposure to the external γ -rays. According to ICRP, effective dose rate is calculated as follows [9]:

$$E = \sum H w_T \quad (2)$$

where H, is the equivalent dose rate and w_T , is the tissue weighting factor for organ or tissue T. Indeed, the configuration of exposure at REEs project represents a whole body exposure. Accordingly, $\sum w_T$ in equation (2) equals unity. Finally the values of the effective dose rate E_γ ($\mu\text{Sv}/\text{h}$) is taken as that of the equivalent dose rate H ($\mu\text{Sv}/\text{h}$). The annual external effective dose E_γ (mSv/y) received by the occupational workers through processing of monazite assuming 2000 working hours per year is calculated as follows:

$$E_{ex} (\text{mSv}/\text{y}) = E_\gamma (\mu\text{Sv}/\text{h}) \times 2000 \text{ h} \quad (3)$$

Table (1) represents the equivalent dose rate H ($\mu\text{Sv}/\text{h}$) while Table (2) shows the external annual effective doses E_{ex} (mSv/y) at the different stages of chemical processes of monazite. From Table (2), clear that all values of the annual effective dose much below the recommended limit of 20 (mSv/y) [9].

Internal exposure:-

- **Effective dose due to radon and thoron gases:-**

Radon has long been recognized as a cause of lung cancer, and was identified as a human lung carcinogen in 1986 by the World Health Organization (WHO) [10,11]. Table (1) represents the activity concentrations of radon and thoron gases. It's clear that the activity concentrations of radon and thoron gases are much below the reported limits of 2000 and 740 (Bq/m^3) for radon and thoron respectively [12]. This is due to the good ventilation at the hanger of REEs project minimizes the concentrations of these gases at the location.

However, many models were established to estimate the effective doses received by the workers due to the inhalation of radon and thoron gases and their decay products. These models depend on epidemiological and

physical consideration [12]. However, the range of dose conversion factors for radon varies from 6 to 15 nSv/(Bq.h.m⁻³). Given this range of values of the dose conversion factor, the established value of 9 nSv/(Bq.h.m⁻³), used in past calculation [13,14] is still considered appropriate for average effective dose calculations [15]. Accordingly, the annual effective doses E_{Rn} and E_{Tn} (mSv/y) received by the workers at the REEs project due to the inhalation of radon and thoron gases and their decay products are calculated respectively as follows [15]:

For radon,

$$E_{Rn} = C_{Rn} (\text{Bq.m}^{-3}) \times 0.4 \times 2000\text{h} \times 9\text{nSv} (\text{Bq h m}^{-3})^{-1} \quad (4)$$

For thoron

$$E_{Tn} = C_{Tn} (\text{Bq.m}^{-3}) \times 0.03 \times 2000\text{h} \times 40\text{nSv} (\text{Bq h m}^{-3})^{-1} \quad (5)$$

Table (2) represents the values of the annual effective dose of radon and thoron gases. From the table, all values of both E_{Rn} and E_{Tn} are much lower than the recommended limit of 20 (mSv/y).

- **Annual effective dose due to U-content**

Intakes of radionuclides can occur via a number of routes. In the case of occupational exposure the main route of intake is by inhalation; a fraction of material deposited in the respiratory system will, however, be transferred to the throat and swallowed, giving the opportunity for absorption in the gastrointestinal (GI) tract. For intakes by inhalation, the ICRP [16] has described a Human Respiratory Tract Model (HRTM), which has replaced the lung model adopted in Ref. [17]. The HRTM takes account of recent information on the physiology of the lungs and is intended to be applicable to the interpretation of bioassay data as well as the calculation of dose coefficients. It has been used for the calculation of inhalation dose coefficients given in the BSS [18]. The current biokinetic models of the ICRP for systemic activity [19-22] were used for calculating dose coefficients given in the BSS [18] for intakes by inhalation and ingestion. The estimated derived air concentration DAC_U for U-238 expected to be inhaled by the occupational workers [23]:

$$DAC_U = I_U / 2000 \times 1.2 \quad (6)$$

DAC_U: derived air concentration for U-238 (Bq/m³),

I_{U,inh,L}: limit of intake of U-238 which is [23]:

$$I_U = L / e(g)_U \quad (7)$$

where L: dose limit, 20 (mSv/y),

e(g)_U: committed effective dose per unit intake (Sv/Bq) = 5.7 × 10⁻⁶ (Sv/Bq) [18].

Using equations (6) and (7), the derived air concentration of uranium DAC_U is 1.46 (Bq/m³).

Table (1) represents the uranium content in the air at the location of extraction of REEs from monazite. It is clear that the uranium contents are below the recommended limit of 1.46 (Bq/m³). From the table (1), the uranium content in the air at the digestion stage is comparable to the recommended concentration of 1.46 (Bq/m³) while it is at the stage of sample preparation lower by one order of magnitude. This is because the time of feeding of the sample into reactor is larger than the time of preparation which allows the dust to get higher concentrations in the air.

The annual effective dose E_U (mSv/y) received by the workers at REEs project is calculated as follows:

$$E_U = C_U (\text{Bq/m}^3) \times 1.2 (\text{m}^3/\text{h}) \times 2000\text{h} \times 5.7 \times 10^{-6} (\text{Sv/Bq}) \quad (8)$$

Table (2) represents the annual effective dose due the inhalation of U-238, E_U, received by the workers at RREs project. The value of E_U (mSv) at the different stages of the chemical extraction is lower than the recommended limit, 20 (mSv). However, technical enhancement is needed to lower the dispersion of the dust into the air of REEs hanger. This may be accomplished by automation of the feeding of the reactor. This justifies the enclosure of the whole process.

Total annual effective dose:-

The total annual effective dose received by the workers at REEs project is calculated as follows:

$$E_{\text{total}} = E_{\text{ext}} + E_{\text{Rn}} + E_{\text{Tn}} + E_{\text{U}} \quad (9)$$

Table (2) represents that total annual effective dose E_{total} (mSv/y) at the different stages of extraction of REEs from monazite. It's clear that the total annual effective doses are within limits since the dose limit for occupational is 20 (mSv/y). The majority of the effective dose comes from the uranium content in the air dust. Also, concentration of U-content at the digestion stage leads to a higher value of the total annual effective dose E_{total} at this stage.

Monazite processing by adding BaCl:-

Table (3) and (4) represent the external and internal exposure through different stages of REEs extraction from monazite and the total annual effective dose by adding BaCl through digestion stage which precipitates radium from the solution. It is expected to minimize the dose rates through the reminder stages. However, all types of exposures are much below the recommended limits. On one hand the radioactive source has a limited total specific activity of 55636 (Bq/m³) [24], or a total activity of 5.6×10^5 (Bq). On the other hand the hanger of the project at NMA is wide enough and has good natural ventilation. This fades the probable effect of adding BaCl at the stage of digestion.

Background:-

Table (5) compares the background of the types of exposure at the REEs project and some laboratories of NMA. It is clear from the table that the background of each type of exposure is at the REEs project is higher than the corresponding type of background at the regular laboratories, comparing table (1) and (5), it is clear that the background concentrations of radon and thoron gases and the background gamma effective dose rate are almost similar to that due to the studied process on 10 kg of monazite. This is due to the REEs project contain some findings that belong to other previous separation patches. Besides, contamination due to the separation process is accumulated on the surfaces of the used instruments and wall and floor of the REEs hanger.

Table (1): Gamma equivalent dose rate H ($\mu\text{Sv/h}$); concentrations of radon gas C_{Rn} (Bq/m³); thoron gas C_{Tn} (Bq/m³); uranium concentrations C_{U} (Bq/m³) at the different stages of flow sheet.

No.	stages	H ($\mu\text{Sv/h}$)	C_{Rn} (Bq/m ³)	C_{Tn} (Bq/m ³)	C_{U} (Bq/m ³)
1	(heating)+sample preparation (weighting)	0.21	14.40	56.88	0.29
2	Digestion	0.11	8.56	56.88	0.88
3	Adjust of pH and pumping the filtrate	0.15	5.86	114.7	BDL
4	filtration of thorium	0.12	0.81	100.5	-
5	Adjust of pH	0.18	8.56	70.98	-
6	washing	0.11	14.15	56.88	-
	Average	0.15	8.72	76.14	0.39

BDL= below detection limit; - = not measured

Table (2): Annual effective dose received by the workers from gamma rays E_{ext} (mSv); radon gas E_{Rn} (mSv); thoron gas E_{Tn} (mSv); uranium concentrations, E_{U} (mSv), total internal effective dose E_{int} (mSv); and total annual effective dose E_{total} (mSv).

stage no	stages	E_{ext} (mSv)	E_{Rn} (mSv)	E_{Tn} (mSv)	E_{U} (mSv)	E_{int} (mSv)	E_{total} (mSv)
1	(heating) sample preparation (weighting)	0.42	0.10	0.14	3.97	4.21	4.63
2	Digestion	0.22	0.06	0.14	12.04	12.24	12.46
3	Adjust of pH and pumping the filtrate	0.29	0.04	0.28	0.00	0.32	0.61
4	filtration of thorium	0.24	0.01	0.24	-	0.25	0.49
5	Adjust of pH	0.36	0.06	0.17	-	0.23	0.59
6	washing	0.22	0.10	0.14	-	0.24	0.46
	average	0.29	0.06	0.18	5.34	2.92	3.21

Table (3): Gamma equivalent dose rate H ($\mu\text{Sv/h}$); concentrations of radon gas C_{Rn} (Bq/m³); thoron gas C_{Tn} (Bq/m³); uranium concentrations C_{U} (Bq/m³) at the different stages of flow sheet with adding BaCl at the digestion stage.

stage no	stages of processing	H ($\mu\text{Sv/h}$)	C_{Rn} (Bq/m^3)	C_{Tn} (Bq/m^3)	C_{U} (Bq/m^3)
1	(heating)+sample preparation (weighting)	0.11	9.10	66.51	0.29
2	Digestion with adding BaCl	0.11	8.90	58.36	0.88
3	Adjust of pH and pumping the filtrate	0.11	10.42	62.07	BDL
4	filtration of thorium	0.12	3.82	94.00	-
5	Adjust of pH	0.31	10.81	58.27	-
6	washing	0.17	11.32	62.54	-
	average	0.16	9.06	66.96	0.39

Table (4): Gamma rays E_{ex} (mSv); annual effective dose received by the workers from Radon gas E_{Rn} (mSv) ; thoron gas E_{Tn} (mSv) ; total internal effective dose E_{int} (mSv) ; and total annual effective dose E_{total} (mSv).

stage no	stages	E_{ext} (mSv)	E_{Rn} (mSv)	E_{Tn} (mSv)	E_{U} (mSv)	E_{int} (mSv)	E_{total} (mSv)
1	(heating)+sample preparation (weighting)	0.22	0.07	0.16	3.97	4.20	4.42
2	Digestion with adding BaCl	0.22	0.06	0.14	12.04	12.24	12.46
3	Adjust of pH and pumping the filtrate	0.22	0.08	0.15	0.00	0.23	0.45
4	filtration of thorium	0.24	0.03	0.23	-	0.26	0.50
5	Adjust of pH	0.62	0.08	0.14	-	0.22	0.84
6	Washing	0.34	0.08	0.15	-	0.23	0.57
	average	0.31	0.07	0.16	5.34	2.90	3.21

Table (5): Background concentrations of radon and thoron gases (Bq/m^3) at the REEs project and other laboratories at NMA. Background value of the annual effective dose rate ($\mu\text{Sv/h}$) is represented.

Type	At REEs	Labs
C_{Rn} (Bq/m^3)	8.72	1.57
C_{Tn} (Bq/m^3)	76.14	6.29
E_{γ} ($\mu\text{Sv/h}$)	0.11	0.07

Recommendation:-

The relative high background at the hanger of the REEs project at the nuclear materials authority, NMA, Egypt, strongly recommends the periodic clearance of the instruments, walls and the floor.

Conclusion:-

Using 10 kg of monazite concentrate (50% purity) to separate REES added an average total effective dose of 3.21 (mSv/y) to the environment of the hanger of REEs project. The wide dimension of the REEs hanger along with the good ventilation faded the effect of adding BaCl at the stage of digestion such that no difference appeared in the total effective dose received by the workers at the REEs. It is recommended to make technical enhancement to reduce the U-content in the air of the REEs hanger.

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