MINISTRY OF EDUCATION AND TRAINING

HANOI UNIVERSITY OF MINING AND GEOLOGY

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STUDY ON ENVIRONMENTAL CHANGES RELATE RADIONUCLIDE DISPERSION CHARACTERISTICS DUE TO MINING AND PROCESSING ACTIVITIES COPPER-ORE AT SIN QUYEN DEPOSIT, LAO CAI PROVINCE

MAJOR: **GEOPHYSICAL ENGINEERING**

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SUMMARY OF DOCTORAL THESIS

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The study was completed at the **Geophysics department, Faculty of Oil and gas, University of Mining and Geology**

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The thesis will be defended at the scientific council of the University of Mining and Geology at…………..

The thesis can be found at the library: **National Library, Hanoi** or **Library of University of Mining and Geology**

Introduction

1. NECESSARY OF THE THESIS

Sin Quyen copper mine, Bat Xat district, Lao Cai province was discovered in 1961 by geologists. In 1969 Geological Team no 5 carried out meticulous exploration and in 1974 completed the exploration works. The mine has an area of 200 ha and has been licensed by the Ministry of Natural Resources and Environment to exploit. Total reserves are about 53.5 million tons of copper ore, average content is 0.95% Cu (including gold, silver, iron, and rare earth elements), uranium content in copper ore is from about 20 to 600 ppm, thorium content about 2 to 20 ppm. Sin Quyen copper mine contains radioactive substances and is considered the largest copper mine in Vietnam.

In the mining and processing of millions copper ores and soils were excavated, transported, stored, removed, crushed, enriched, etc. Those materials contain high radioactive substances, causing radioactivity release strongly into the surrounding environment, especially dispersed in water and air. Dust containing radioactive material can be carried by the wind to residential areas and areas located far from the mine. Impact due to mining and processing of radioactive copper ore mines can change the amount and dose of radiation expose.

However, current domestic studies have not paid attention to the mining and processing of radioactive ores, and there is no method to properly assess the radioactive environmental effects caused by mining activities in officially. Those are all of reasons why have this study.

2. OBJECTIVE AND STUDY SCOPE

- Objective: radioactive substance's in Sin Quyen copper mine, Lao Cai province
- Study scope: Ore mining and processing area and residential areas adjacent to Sin Quyen copper mine, Lao Cai.

3. RESEARCH PURPOSES

- Characteristics of radioactive substance's dispersion leading to changes in radiation concentration and dose due to mining and processing activities at Sin Quyen deposit, Lao Cai.

- Assessment of the radiation effects on the environment and human health due to the mining and processing of radioactive copper ores in the Sin Quyen, Lao Cai.

4. RESEARCH CONTENT

- Collecting and synthesizing documents and research works on the investigation and assessment of the radioactive environment, the characteristics of radioactive substances dispersing into the environment to change the concentration and dose of radiation.
- Collect and synthesize documents on geology, minerals, geo-chemistry, and environment ... so far in the study area.
- Studying the geo-environment characteristics, distribution and existence form of uranium in order to elucidate the geochemical characteristics of radioactive substances in the process of mining and processing copper ore.
- Study on characteristics radionuclides (in soil, water and air environment).
- Research and build models from theory to experiment to determine the radon release.
- Researching methods of statistical analysis determine the local natural radiation background before mining and processing and the current dose after mining and processing as a basis for determining the fluctuation of radiation dose assessment of environmental pollution.
- Evaluation of the effects of radioactivity on the surrounding environment due to copper ore mining and processing at Sin Quyen deposit, Lao Cai.

5. RESEARCH METHODOLOGY

To carry out the above research, the researcher used a combination of research methods, including:

- Research on the theoretical basis of radioactive release characteristics due to the mining and processing of ore bearing radioactive minerals in the world and in Vietnam.
- Methods for building theoretical and practical models to predict radon concentrations in the environment when there is mining and ore processing.
- Method of determining the change in radioactivity content and dose when mining and processing radioactive ores.

- Methodology for interpreting calculation and measurement results in order to select appropriate methods and to assess radioactive environmental impacts caused by mining and mineral processing activities.

6. METERIALS

The thesis is made on the basis of documents collected by postgraduate at the radioactive environment during working time at office. The geological, geochemical and environmental documents of scientists researching on Sin Quyen copper mine, Lao Cai, the author has directly constructed the monitoring, investigation and assessment of the radioactive environment at study area. In addition, author also consulted the documents of many domestic and foreign research projects related to radioactive studies relating the radioactivity effect due to when there are mining and mineral processing activities...

7. NEW POINTS OF THE THESIS

7.1. The new research methods are based on approaches that have been introduced in the world: author study the geochemical environment characteristics of the Sin Quyen copper mine under the influence of mining and processing activities, and detailed survey methods of the radioactive environment to determine the radiation dose distribution in mining and processing areas and surrounding residential areas.

7.2. The author were distinguished and clarified the geochemical environmental characteristics of the mining and processing areas at Sin Quyen copper mine. The characteristics of radioactive dispersal at Sin Quyen copper mine in the aquatic environment are closely related to the geochemical and ore-chemical characteristics namely radioactive minerals, including uranitite.

7.3. Building a model from theory to experiment determine the dispersion of radon gas release along low-terrain valleys and accumulate in residential areas with many houses and trees to block the wind.

7.4. The results of assessment of radioactive environmental effects due to mining and processing of copper ore at Sin Quyen deposit, Lao Cai which can serve population planning and socio-economic development.

8. THE PROTECTION THESIS

8.1. Thesis 1: The methodology of surveying the radioactive environment was combined with sampling for geological and geochemical analysis that is a reasonable and reliable research method to help clarify the characteristics of the environment. The environment in the mining and processing areas are convenient for the dissolution and transportation of radioactive substances

8.2. Thesis 2: The characteristics of radioactive dispersion in the water environment due to the exploitation and processing of copper ore, Sin Quyen deposit are determined as follows:

+ When exploiting, the water at the field site has a high content of HCO_3- anion (from 30 to 292 mg/l, average 125 mg/l) and pH from 6.3 to 8.75, average 7. 3 features a weak alkaline environment that could increases the solubility of uranium from the solid mineral uraninite dozens of times.

+ When processing copper ore, pyrite and other sulfide minerals are crushed and mixed with the ore to create a sulfuaric acid environment that reduced the pH from 7.3 to 2.7 (acidic environment) and strongly increases solubility of uranium up to hundreds of times.

9. SCIENTIFIC AND PRACTICAL

* Scientific significance:

- The research results of the thesis full fill and more comprehensive awareness of radioactive dispersion characteristics in Sin Quyen copper mine.
- The research results of the thesis contribute to perfecting the methodological system to study the characteristics of radioactive release and environmental changes caused by mining and processing not only in Sin Quyen copper mine but also applied to other mines with similar conditions.
- Determining the change in the radioactive environment helps to assess the radioactive environmental effects due to mining and processing of copper ore bearing uranium, Sin Quyen deposit.

* Practical significance:

The accurate determination of changes in the radioactive environment in the Sin Quyen copper mine area, Lao Cai which is based on the actual survey measurements in the field, the sample analysis data at the laboratories at

AGH, Poland and the lab had high reliability to help accurately assess radioactive environmental impacts, for planning of exploitation and processing of copper ore from Sin Quyen deposit and population planning and socio-economic development.

The thesis was completed at the Department of Geophysics, HaNoi University of Mining and Geology under the scientific supervisor of Prof. Dr. Le Khanh Phon, and Prof. Dr. Nguyen Van Lam. The postgraduate expressed his deep gratitude to the scientific instructors who were always closely and devotedly throughout the process of studying, researching and completing the thesis. In the process of implementing the thesis, the postgraduate always received the attention and help and created favorable conditions from the Department of Geophysics, the Faculty oil and gas, the University of Mining and Geology, postgraduate department, Ministry of natural resources and environment, General Department of Geology and Minerals of Vietnam, Geology Division for Radioactive and Rare Elements, scientists, and colleagues. I appreciate it!

CHAPTER 1. OVERVIEW

1.1. Research situation on the dispersion of radioactive substances due to mining and processing in the world

In the world, many studies òn the dispersion of natural radioactive substances in the environment caused by the influence of the processes of mining and processing radioactive ores and containing radioactive materials. Fernandes and collaborators (Brazil) [50], Jenk and Schreyer (Germany) [51], Chrusciel [52], Pieczonka and Piestrzynki (Poland) [53], Carvalho et al (Portugal) [55] [58], Tripathi et al (India) [56] and many other scientists around the world have conducted studies to identify and evaluate the level of danger by increasing levels of radioactive elements in environment. Environmental radiation due to uranium and radium ore mining. The results of the survey and assessment have given measures to restore the environment and measures to ensure radiation safety for the surrounding environment and people.

Not is only studying the radioactive environment at uranium mines, other bearing radioactive deposits are also interested. Roxman and others (Russia) [74], Adagunodo et al [61], Gaafar et al (Egypt) [59] studied and determined the elevation of radioactivity in copper-molybdenum deposits, kaolin mines... thereby providing requirements for radiation protection in those radioactive mines.

To clarify the radioactive dispersion in the environment during the extraction and processing of radioactive minerals, Roxman GI, Bakhur AE, Petrova NV (Russia) [74] divided radioactive wastes in liquid form, gas, and solid.

a. Dispersion of radioactive substances in liquid form:

b. The release of radioactive gaseous substances:

c. Dispersion of radioactive solids:

* Comment: In the world, in order to assess the radioactive environmental effects caused by mining and processing radioactive ores, two approaches have been proposed:

- The first approach: study the geochemical environment characteristics of mineral deposits in the oxidizing conditions of the epigenetic zone.

- The second approach: applying a detailed surveying method of the radioactive environment, determining the rules of distribution of radiation dose concentrations at the field, flotation plant, waste dump and neighboring residential areas.

1.2. Research overview in Vietnam

In the study of the characteristics of the dispersion of radioactive substances in radioactive mines, there have been many studies, such as Tran Binh Trong [23, 26], Nguyen Van Nam [23, 26], […] 28], Trinh Dinh Huan [31, 35], Nguyen Phuong [36], Nguyen Thai Son [37]. Most studies on the characteristics of radioactive substance's dispersion in the natural environment. In the field of investigation and assessment of environmental impacts caused by mining and processing activities of radioactive and radioactive minerals in Vietnam, they have not been paid much attention and studied in detail. Specifically, only the study by the group of authors Trinh Dinh Huan [31] initially provided the data as evidence about the variation of irradiation dose and the risk of radioactive pollution caused by the activities of the uranium ore exploration in the area of A block, uranium mine in Pa Lua - Pa Rong area, Nam Giang district, Quang Nam province. However, the study did not calculate the variable dose and clarify the dispersion characteristics of radioactive substances in the environmental components, so it did not fully assess the effects of the radioactive environment on the staff and the population in the vicinity of the surrounding mine.

1.3. The shortcomings and research tasks of the thesis

As mentioned by the author in the above section, the scientific works conducted by agencies, localities and scientists in our country have only mentioned the investigation, survey and assessment of the environmental impact field natural radiation. The methodology and research directions on the dispersion characteristics of radioactive substances that change the environment due to mining and processing activities which have not yet received attention, research and development tasks to have a clear and scientific view on effects of radiation in the process of mining and processing radioactive ores, containing radioactive materials in our country. Therefore, the tasks of postgraduate needs to solve the following problems:

- Identify the geochemical environment and dispersion characteristics that change the irradiation dose concentration due to mining and mineral processing activities.
- Develop a system of methods for assessing the radioactive environment in ore mining and processing activities, clearly defining the concept of "radiation work" (human activities that change radiation levels and doses) such as exploration, extraction and processing of radioactive minerals).
- Clarifying the concept of "natural radiation background" when assessing radioactive effects on "radiation work" is "local natural radiation background" which is determined on the area of the human impact change in the dose, irradiation dose.

CHAPTER 2. GEOLOGICAL CHARACTERISTICS, OVERVIEW OF MINING AND PROCESSING AT SIN QUYEN COPPER MINE, LAO CAI PROVINCE

2.1. Location and history of geological and geophysical research in Sin Quyen deposit, Lao Cai province.

2.1.1. Overview of the location of the research area of radioactive copper mines

Sin Quyen copper mine belongs to the right bank of the Red River side, extending from Vi Kem and Coc My villages to the center of Ban Vuoc commune, Bat Xat district, Lao Cai province. The mine area covers an area of 200 ha and is licensed by the Ministry of Natural Resources and Environment.

2.1.2. History of geological and geophysical research in Sin Quyen deposit area

In 1960, Delegation 135 discovered radioactive abnormalities near Vi Kem, Sin Quyen villages. In 1975, the results of meticulous exploration of Sin Quyen copper mine identified high radioactive content in copper ore bodies using borehole geophysical documents, showing a high correlation between copper ore bodies and strong measured gamma levels, proving that there is a radioactive symbiosis in Sin Quyen copper ore.

Mining work in Dong Sin Quyen - Lao Cai has been conducted by Vietnam Minerals Corporation since 2006 until now.

In the 2000s, Le Khanh Phon carried out investigation and the survey of the radioactive environment in the area, the results determined that some components of the radioactive environment in the mine area exceeded the allowable limit.

2.2. Natural and socio-economic characteristics related to Sin Quyen deposit

Sin Quyen area is located in the northeast wing of Hoang Lien Son range and in the southwestern of the Red River zone. The hills here are stretched into strike in the direction of Northwest - Southeast. The area is very complicatedly dissected, the valleys in this area are wide, and the mountains are steeps.

- Climate characteristics: Sin Quyen deposit area is located in the northwest of our country, so it has the general characteristics of a tropical climate. In the cold season, it is influenced by the Northeast monsoon, and in the summer, it is influenced by a very hot and dry. The climate in the region is divided into two distinct seasons: the dry season starts from October to April next year, the rainy season from May to September. The annual average temperature is 22.50C, the highest is 420, the lowest is low at least 20. Humidity from 70 to 85%. The annual rainfall is from 1400 to 1800 mm, the maximum daily rainfall is 212 mm.

- Hydrological features: Rivers and streams in the region flow in two directions almost at right angles to each other, in the northwest-southeast direction and a system of streams flows in the southwest-northeast direction. The Red River is the only and largest river in the region.

2.3. Geological and mineral characteristics of the study area

2.3.1. Stratigraphy

In the study region there are sedimentary and metamorphic formations of Proterozoic, Paleozoic and also Cenozoic era according Ta et al, (1975).

Proterozoic

The Sin Quyen deposit occurs in Proterozoic Sin Quyen formation (SqF). The SqF includes sedimentary rocks and metamorphosed sediments hosting copper bodies. The formation is composed of highly crushed amphibolites, migmatised gneiss, granite-gneiss, schist and altered rocks extending in NW–SE direction between 280 and 320°, 50 to 85° of dip, and 400-800m of thickness. The SqF underlies comformable with the Thung Sang formation (TSF) and Cambrian Sa Pa Suite (SPS), and uncomformable with Cam Duong formation (CDF), and Cenozoic Quaternary formations (Q). The structure of the mentioned formations were strongly controlled by Sin Quyen reverse fault and also were penetrated by number of regional dislocations, including Sin Quyen, Pin Ngan Chai and Thung Sang faults, all of them are parallel and visible as splays of the major Red River Fault (RRF).

From the mineral composition point of view, the SqF can be divided into lower (Sq1) and upper (Sq2) sub-formations. The Sq1 contain 50% of quartz, 15% of graphite, 10% of biotite, and 12% of muscovite as a major minerals and plagioclase, tourmaline, garnet, sillimanite as a minor minerals. While the Sq2 is chiefly consist 61% of plagioclase, 21% of quartz, and 15% of biotite as major minerals and apatite, sphene, calcite and garnet as accessory minerals.

Paleozoic

The CDF is composed of Paleozoic Cambrian sediments composed of quartz, sericite, schist containing graphite, carbonate, schist, quartz, biotite, chlorite schist. The formation is extending in NW-SE strikes from 280 to 320° and dipping under 20 to 70°.

Cenozoic

Cenozoic Quaternary sedimentary formations (Q) in the region are very thin and uncommon, and consist of pebble, sand, clay and sandstone.

2.3.2. Intrusives

The magmatic intrusives are divided into two main formations: Proterozoic Coc My formations (CMF) and Permian suites. The CMF covers up to 30% of the deposit area and consists of amphibolite and granite-gneiss. Permian intrusives are the youngest in the region, and are represented by gabbro–

dolerite and plagio-granite intrusions Ta et al, (1975).

Amphibolites are the earliest intrusive rocks penetrating into the SqF with thickness from 5 to 50 m and 10 to 100 m in length, extending in NW–SE direction. The intrusive rocks comprise 66% of hornblende, 19% of plagioclase, 6% of biotite as major minerals and other minor minerals such as orthite, epidote, apatite, ores, chlorite and calcite. In the zone close to the copper ore bodies there usually occur other minerals such as allanite, calcite, and quartz.

Granite-gneiss occupies up to 25% of the deposit and is highly crushed and migmatized. Those rocks occur as dykes and lenses 2-200 m in thickness, 10-3000 m in length and extend in NW–SE direction. The granite-gneiss comprises 66% of plagioclase, 26% of quartz, and 7% of biotite as major minerals and zircon and apatite as minor minerals.

The gabbro–dolerite occurs in the zone of contact between the SqF and the SPS and extends in NW–SE direction. The gabbro–dolerite comprises of amphibole [64%], plagioclase [18%], biotite [7%], chlorite [5%] and other: calcite and disseminated pyrite, pyrrhotite, chalcopyrite.

The plagio-granites occur in fracture zones of the deposit and penetrate through the SqF into SPS rocks and the CMF with 0.5-20 m thickness and 10-300 m length. The plagio-granites consist of plagioclase (63%), quartz (26%) and biotite (6%) as major minerals and microline, muscovite, apatite, chlorite, and sometimes traces of zircon and albite as minor minerals. In the zone close to the plagio-granite there is plagio-pegmatite composed of quartz, plagioclase, biotite, allanite and epidote.

2.3.3. Constructive characteristics

The metamorphic formations were briefly described by Ta et al, (1975). They occur at the center of deposit in various forms such as lenses, chambers or vein bunches with 0.5 – 100 m thick, 1 - 1000 m long and trend from 280^0 to 320^0 and are dipping from 65^0 to 90^0. The metamorphic formations contain pyroxene, garnet, hastingsite, quartz, albite, orthite, apatite, biotite, plagioclase, vezuvian, chlorite, epidote, calcite and allanite. The metamorphic rocks can be divided into two groups: 1) skarn-pyroxene-garnet rocks and 2) skarn-hastingsite-biotite-quartz rocks groups. The skarn-pyroxene-garnet rocks group composed of pyrrhotite ores, rare chalcopyrite and the skarn-hastingsite-biotite-quartz group including magnetite ore, uraninite, sulfide ore (mainly chalcopyrite-pyrrhotite).

2.3.4. Resource

The Sin Quyen deposit is the largest IOCG deposit in North Vietnam, with a total area above 100 ha and includes seventeen ore-bodies. The reserves of Cu, REE, S, Au and Ag amount to 550 000; 334 000; 843 000; 34.7 and 25.3 tons, respectively. Since 2006, the Sin Quyen deposit has been exploited as an open pit mine with an annual average production of 12,000 tons of copper metal. Every year, more than million tons of ore and seven million cubic meters of rocks are excavated.

CHAPTER 3. CHARACTERISTICS OF RADIOACTIVE DISPERSION IN SINQUYEN DEPOSIT, LAO CAI

3.1. Research method

3.1.1. Scientific basis for selecting research methods

3.1.1.1. Dissolution of uranium from ores, minerals and rocks in natural water

a. Anion composition in water effect conversion of uranium from the ore into aqueous solution: The presence of HCO3- ions in water greatly increased the amount of uranium transferred from the ore into the solution. When even pH = 7 the content of uranium in the solution containing HCO_3- ions increased tenfold.

b. Effect of oxygen on the displacement of minerals and ores and aqueous solutions: According to experimental documents [70],[74], it was shown that, when dissolving the minerals uraninite in oxygen-containing water with the presence of pyrite, which creates a sulfuric acid environment in solution, greatly increases the solubility of uranium in the mineral, especially when the water is saturated with oxygen. From pyrite-containing minerals, in the presence of water containing oxygen, the amount of uranium in aqueous solution is 20 times greater than that of no oxygen (see table 3.1).

c. Effect of the material composition of the ore on the conversion of uranium into aqueous solution: In the acidic sulphate environment, uranium is converted more strongly than in the carbonate environment. In the presence of pyrite alone, the uranium content in solution increases 100 fold even at pH = 4.5.

3.1.1.2. Transport forms of uranium in water

The forms of uranium transport in water are varied and depend on the pH and chemical composition of the water.

3.2. Methods to study radioactive material dispersion caused by mining and processing at Sin Quyen copper deposite

3.2.1. Methods of c***o***llecting and synthesizing documents:

+ Pre-exploitation survey.

+ Survey year 2000.

+ Survey of mining and processing companies.

+ Survey of radioactive environment has been carried out within the framework of the bilateral cooperation project between Vietnam and Poland.

3.2.2. Environmental geological route method

3.2.3. Method of measuring environmental gamma dose rate

3.2.4. Environmental gamma spectroscopy method

3.2.5. The method of alpha spectroscopy measures radon and toron concentrations in the air

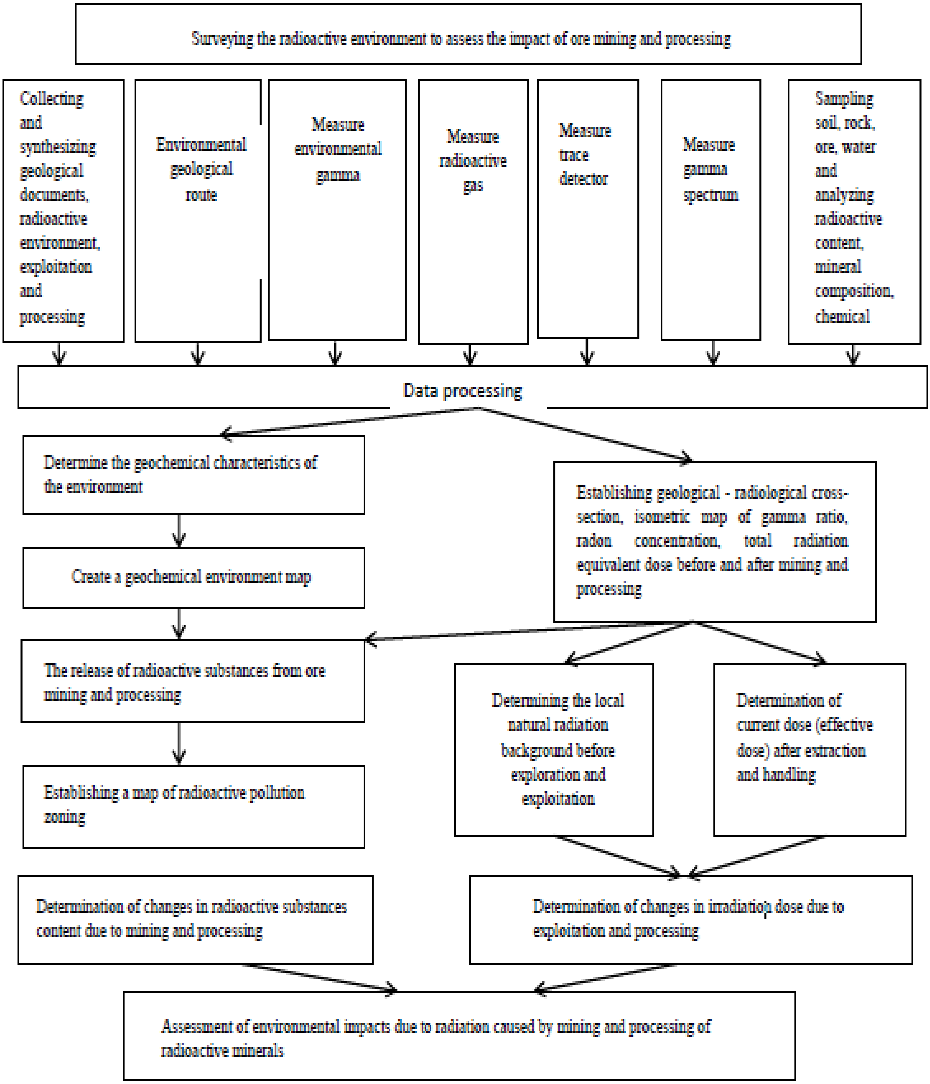
3.2.6. Method of measuring alpha trace detector

3.2.7. Methods of monitoring radioactive environment

3.2.8. Sample collection and analysis

3.2.9. Methods of analyzing, processing and synthesizing documents

A summary of the research methodology is as follows:



3.3. Research results on dispersion characteristics due to copper ore mining and processing, Sin Quyen deposite

3.3.1. Environmental characteristics of the Sin Quyen copper mine area

The radioactive environment in the study area includes the "background" radioactive environment before mining and the radioactive environment (temporarily referred to as the post-exploitation radioactive environment).

3.3.1.1. Features of mineral composition, chemical composition of rock, copper ore and waste in Sin Quyen area:

The results of microscopy in 40 samples of rock, ore and waste have determined the mineral composition in Sin Quyen Lao Cai area, including the main minerals magnetite, pyrite, pyrotin, chalcopyrite, sphalerite; ilmenite, marcasite, tennantite, cubanite, arsenopyrite, galena, bismuth are minor minerals. Microscopic results have also identified crystals of Uraninite in magnetite - large sulphide ores

The results of gamma spectrometry in the laboratory of isotopic compositions ^{238}U, ^{232}Th, ^{40}K in rock and ore samples at Sin Quyen copper mine have confirmed the correlation coefficient of 0.78 between ^{238}U and copper. Thus, in the Sin Quyen, there is a close relation between ^{238}U with copper ore.

3.3.1.2. Characteristics of geochemical environment in Sin Quyen copper deposit area

The results of the chemical composition analysis of the samples showed that the water at the quarry in the Sin Quyen deposit area had a high content of HCO_3^- anion from 82 to 272 mg/l, the average was 178 mg/l, the pH fluctuated between 82 and 272 mg/l range from 7.2 to 8.05 with average 7.7 which is characteristic for weak alkaline environment. Besides, the E_h redox potential of the water in the mining area all have Eh values > 250mV, which is typical for a strong oxidizing environment which is a favorable environment for dissolving uranium from its minerals into the environment. On the geochemical environment map, two anomalies of ^{238}U and ^{234}U have been identified in the West and the East sides. The anomaly ^{238}U, ^{234}U in the western has the form extending from the southwest to the northeast in the direction of the flow from the west field to the water sampling location at Ngoi Phat. This proves that the uranium in copper ore due to mining is exposed in the aeration zone, is dissolved, transported, and released into the water of Ngoi Phat.

In the area of flotation plant, during the processing, rocks and copper ores containing pyrite and other sulfide minerals are crushed and mixed. Under the effect of oxygen and natural water, sulfur is oxidized to create a sulfuric acid environment, causing the pH in wastewater to change rapidly, decreasing from 7.3 to the lowest value of 2.7. In the area of the waste lake and lake used for ore extraction, the pH ranges from 2.7 to 3.64, the concentration of HCO_3^- anion is very low below 0.5mg/l in value. The low pH value and strong oxidizing properties (E_h value > 300mV) of the medium increase the solubility and dispersal of uranium from the solid mineral uraninite into the water of the refinery and waste lake. On the geochemical map of the environment, the anomaly ^{238}U with a concentration of 12.7Bq/l and ^{234}U with a concentration of 13.1Bq/l in the waste lake was identified, caused by wastewater with a high concentration of Uranium flows out of a copper ore processing plant.

3.3.2. Dispersion characteristics of radioactive substances in the aquatic environment

When the mine was exploited on a small scale (in 2000), the mining area and the selection plant were located in the western mining area of the mine. Sin Quyen copper mine has not yet built an ore processing area. At that time, the total alpha and beta activities of water samples in the field, mill water, and Ngoi Emission spring water all increased and exceeded the allowable standards (water samples with total activity α > 0.1 Bq/l, total β activity >1.0 Bq/l) which are causes a pollution area of approximately 0.55km^2 covering the entire mining pit and flotation plant.

Since 2015, the scale of the mine has increased, has expanded the copper ore mining area, expanded the West side, put into operation the East side, the site is located more than 1km from the field site, the wastewater lake and the waste dump are located in near those areas.

The survey results of water environment in the mining area, $HCO3^-$ anion content is from 82 to 272 mg/l, average is 178mg/l, average pH is 7.7, which is typical for weak alkaline environment. Eh of the water in the mining area all have E_h>250mV, which is typical for a strong oxidizing environment, and is favorable for dissolving and transporting uranium from minerals to the environment. Therefore, this is the reason why the water in the mining area including the East and West sides and a part of Ngoi Phat stream that crosses the field has a high total activity of α and β exceeding the allowable standard with a total area of about 1.5 km^2, 3 times larger than the polluted area in 2000.

3.3.3. Dispersion characteristics of radioactive substances in the soil environment

Figure 3.9 was established according to the survey results in 2000, when the mining scale was small on the eastern side, the area of radioactive contamination in the soil environment had a uranium content of > 30ppm (exceeding the standard permitted for construction materials) with an area of about 0.4 km^2 located on the Western mining area and the waste dump. Mineral processing area has not been polluted because it has not been built.

Figure 3.10 established according to survey results in 2015 and re-examination in 2017, 2018 shows that, as the scale of ore mining and processing increases, the total area of land pollution (q_u > 30ppm) in the whole the mine area increased nearly 4 times (approximately 1.5km2), of which the area of soil pollution in the mining area (Western, Eastern mining, waste dump) is 1.3km2 and the mineral processing area (workshop) selection and training) is 0.2km^2.

The increased area of soil pollution due to copper ore mining is due to the expansion of Sin Quyen copper mining area. However, the area of radioactive contamination of the soil environment is only located in the mining area, flotation plant, and disposal site, proving that radioactive substances are dispersed in the solid phase. The content of radioactive substances changes in the soil environment because the tailing of ore and waste rock are released, transported around the mining site and stored in the waste dumps.

3.3.4. Characteristics of radioactive release in the air

3.3.4.1. Theoretical and practical basis of method selection

The M5P algorithm was use as a data processing and analysis tool, thereby building a Radon diffusion model based on the specific object of uranium (with characteristic value of gamma dose rate at an altitude of 1m) in Sin Quyen deposit. The dataset used for the dispersion model includes 5,000 network data points with the coordinates of gamma dose rate values in the Sin Quyen copper mine area; data on measuring dose rate of gamma, radon by CR-39 inside and outside the houses of 21 houses around Sin Quyen deposit.

The results after running the model have predicted the concentration of radioactive gas radon in 21 houses in the resettlement area near the mine site. Using M5P build a gas dispersion model with the results of Peason's reliability assessment (Sig < 0.01) on the correlation constant and with the correlation constant value R=0.95 for the difference between mean value of predicted and actual value of cumulative radon activity measured by CR-39 (see figure 3.15).

Therefore, it can be confirmed that the concentration of radioactive radon gas measured in residential areas is due to the emission from the Sin Quyen copper mine during ore mining. However, because only 21 independent houses have been studied at different locations in a resettlement area of Sin Quyen copper mine, the study on radioactive gas emission characteristics in the thesis is only constructive method building and initial testing.

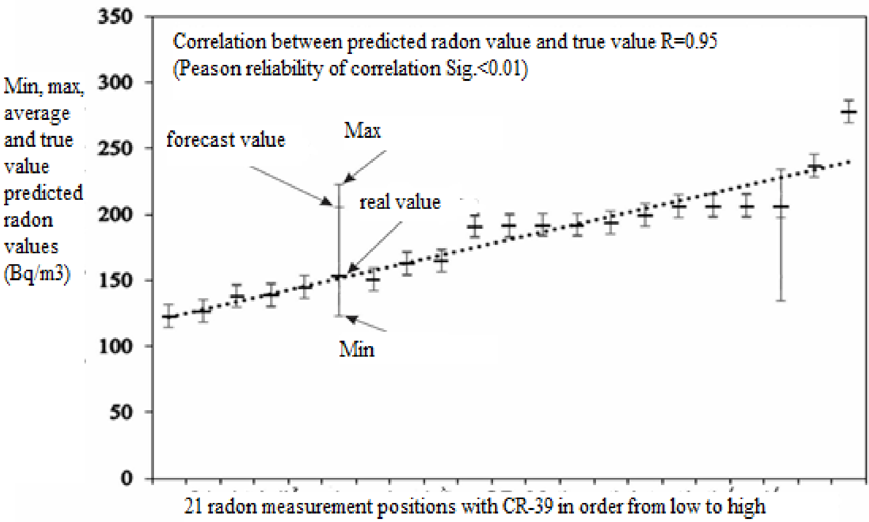


Figure 3.15. The graph shows the predicted correlation of Radon gas emission at houses surrounding the Sin Quyen copper deposit

CHAPTER 4. DETERMINATION OF CHANGE AND ASSESSMENT OF THE RADIATION ENVIRONMENTAL EFFECTS DUE TO MINING AND PROCESSING OF COPPER ORE BEARING URANIUM AT SIN QUYEN DEPOSIT

4.1. Study methods

4.1.1. Selection basis and determination method

4.1.1.1. Basis of method selection

For radiation work, the dose limit value has also been determined by a specific number for A group professionals at 20mSv/year, for C group civilians it is 1mSv/year (not counting natural radiation background). People in the process of living and working can be present at any location in the mine area and subject to the average radiation dose of the entire environment.

4.1.1.2. Method for determining variable dose

In each plot, a graph of gamma radiation dose frequency and radon concentration in the air were built. For the histogram plots with normal distribution, the material composition is considered to be homogenous. Then determine the average value of gamma dose rate, Radon concentration according to the mean value. For cells with heterogeneous composition, the histogram does not have a normal distribution, dividing each plot into 2 parts: the area of cells in the ore body area and the area of cells outside the ore body. Calculate the average value of gamma dose rate and radon concentration of the whole mine before and after exploration, exploitation, and processing. From there, determine the internal dose through inhalation and the external dose.

4.1.2. Determination of changes in radioactive environment due to mining and processing of copper ore from Sin Quyen deposit

4.1.2.1. Determination of radioactive content change

a. Variation of radioactive content in the aquatic environment

The mining and processing process of Sin Quyen copper mine since 2000 has caused a polluted area of approximately 0.55 km^2, covering the entire mining pit and the adjacent smelter and when the scale is large exploitation and processing increased, all water samples at the East and West mine sides, the selection solid waste dump, and the wastewater lake with an area of about $1.9km^2$ more than 7 times larger than the initial survey results.

b. Variation of radioactive content in soil environment

Before mining, the concentrations of radioactive elements U, Th, and K on alluvial layers in Ngoi Phat area were q_U ~ 5ppm, q_{Th} ~ 10ppm, q_K ~ 3%, respectively.

The survey results in 2000, the area with high uranium content q_u> 30ppm about $1km^2$ is located on the mining area, waste dump, flotation plant. From 2015, when the scale of ore mining and processing increased, the area of soil pollution with $q_U \geq 30$ppm increased about 4 times (approximately $1.5km^2$), located on the area of the East and West mining areas. The uranium contents in copper ore varies from 300 to 740 ppm and in rock from 30 to 600 ppm. Thus, the mining and processing activities of Sin Quyen copper ore have changed the radioactive composition in the soil both in terms of scale and content.

c. Variation of radioactive gas concentration

Before mining, the concentration of radon in the air in the study area was low, ranging from 10 to $70Bq/m^3$, the area with concentrations above 30Bq/m3 concentrated in the western side, the inside mine area. This is safety level. In 2000 (Figure 4.2), an area of about $7000m^2$ with a concentration of N_{Rn} > 150 Bq/m^3 exceeded the allowable standard. Survey results in 2015 and inspection results in 2017, 2018 (Figure 4.3), the area of radioactive gas contamination with N_{Rn} > $150Bq/m^3$ was covers the West and East sides, recruitment and residential areas along Ngoi Phat and the right bank of the Red River side with area of tens square kilometers. Since 2015, the area contaminated with radioactive gas has increased 5 times compared to 2000, the concentration of radon has increased approximately twice in comparison with in 2000.

Rn gas at the field site and waste dump spreads in the direction of the wind, spreading to residential areas, more than 1 km away from the mine site, that is the causing an area of radioactive radon pollution exceeding the allowable concentration standards. Because the residential area has low terrain and sheltered by houses and trees, the concentration of radon gas tends to increase gradually from the mine side to the residential area.

4.1.2.2. Determination of irradiation dose variation

a. Determination of gamma dose rate variation

Before mining, the gamma dose rate varied from 0.1 - 0.5µSv/h. Survey results in the 2000s showed that gamma dose rate values varied from 0.1 to > 0.7 µSv/h. The gamma dose rate value has changed significantly in the western side. Since 2015, the dose rates value continues to change significantly in the East and West side, forming an anomaly in the northwest - southeast direction with intensity > 0.3µSv/h including. The characteristics of gamma dose rate in the mine area varies from 0.2 to >1µSv/h, the gamma dose rate ranges from 0.5 to >1µSv/h, the smelter > 0.3µSv/h.

b. Determination of irradiation dose variation

To determine the dose variation during mining and processing, it is necessary to determine the pre-mining irradiation dose and the current irradiation dose (at the time of assessment). Because survey lines and measuring points are often unevenly distributed over the area, the author divided the area of the mine into 74 equal-sized plots, (~0.18km^2 each).

Postgraduates divided the study area into two areas (Figure 4.7) which are residential areas (zone C) and staff areas (zone B), which was used to calculate the effective dose for each specific object.

In each plot, the characteristic value of gamma radiation dose rate and radon concentration in the air are determined by the method of building a histogram.

* Characteristics of pre-exploiting radioactive environmental parameters: Value of gamma dose rate < 0.20 µSv/h occupies about 70% of the area; gamma dose rate values from 0.20-0.30 µSv/h account for about 30%. Therefore, postgraduate determined the value of gamma dose rates before mining as Ig = 0.15x0.7+0.25x0.3=0.18 µSv/h.

Similarly, the concentration of radon gas in the air of the mine area before the mining and processing stage ranges from 10 to 70Bq/m^3 (Figure 4.1), the average value of radon gas concentration before mining is 15x0.7 + 25x0.3 = 18Bq/m^3.

* Characteristics of radioactive environmental parameters after mining and processing

The mine area is divided into two areas: The mine and flotation plant areas (including 49 cells) do not allow people to enter, which means that the radiation dose affects only officials and workers involved in production; area of residential area, waste dump and waste lake (including 25 cells) irradiation doses directly affects the population.

* For the mine and flotation plant areas: build a dose frequency chart (Figure 4.10), determine the specific dose rate value for the field, the flotation plant $\overline{H_{SL}}$ = 0.38 µSv/h.

* For the residential, waste dump and lake areas are divided into 26 cells:

According to the dose frequency chart (Figure 4.11), the average dose rate value for residential areas, dumping sites and waste lakes can be determined: $\overline{H_{SL}}$= 0.22 µSv/h.

- Determine the value of radioactive gas concentration of radon after extraction:

Characteristic value of Rn concentration, N_{Rn} flotation plant = 37.7Bq/m3 (Figure 4.14)

The typical value of Rn in residential areas located on high and open terrain, waste dumps and lakes N_{Rn} = 23.6Bq/m^3 (Figure 4.15).

* Value of total dose before mining and processing (natural radiation background typical of the mine) and dose after mining and processing are determined by the following formula:

$H\sum(mSv/year)$ = Hn(mSv/year) + Hp(mSv/year) + Ht(mSv/year)

Variable dose = $H\sum(mSv/year)$ (after) - $H\sum(mSv/year)$ (before)

On the basis, the characteristic values, the author calculated the dose before and after mining and processing and the variable dose in the study area given in table 4.7:

Table 4.7. Calculation results of variable dose due to mining and processing activities

TT	Dose / Object	Irradiation dose before exploitation and processing (local natural radiation background) (mSv/year)				Irradiation dose after mining and processing (mSv/year)				Variable dose due to mining and processing activities (mSv/year)
		Hn	Hp	Hd	HΣ	Hn	Hp	Hd	HΣ	
1	staffs and employees	1,66	1,00	0,06	2,72	3,33	1,77	0,1	5,2	2,48
2	Resident	1,66	1,00	0,06	2,72	2,00	1,11	0,1	3,21	0,49
3	Resident in the resettlement area	1,66	1,00	0,06	2,72	2,00	6,25	0,1	8,35	5,65

Thus, the variable dose for staffs and workers involved in mining and processing of Sin Quyen copper deposit is 2.48mSv/year, and for scattered resident in high terrain areas, open air is 0.49mSv/year, population in the resettlement area is 5.65mSv/year.

4.2. Assessment of radioactive environmental effects due to copper ore mining and processing at Sin Quyen deposit, Lao Cai

4.2.1. Legal basis for assessment of radioactive environmental effects due to mining and processing of radioactive minerals

The legal basis for the assessment includes the recommendations of the International Commission on Radiological Protection (ICRP):

- Current annual dose level ≥ 10mSv/year is the dose level needed for intervention (start considering intervention actions).
- Current annual dose level < 10mSv/year does not really require intervention. However, interventions can be made to reduce a large proportion of the total dose.
- In cases where the equivalent annual dose level exceeds the dose threshold causing deterministic effects for a certain organ of the body, intervention is necessary.
- If the current annual dose level reaches 100mSv, then intervention is required.

* For the control and assurance of radiation safety in occupational and public irradiation, the provisions of Circular No. 19/2012/TT-BKHCN dated November 8, 2012 of the Ministry of Science and Technology.

4.2.2. Assessment of radioactive environmental effects due to copper ore mining and processing at Sin Quyen deposit, Lao Cai

4.2.2.1. Evaluation of the effect of changes in radioactive content

According to the results of analysis of the concentration of radioactive substances in water samples at fields, flotation plant, waste dumps, and wastewater lakes with total α, β activities exceeded the permissible safety standards, they cannot be used as drinking water activities, eating and drinking (table 4.9).

Rn gas released from mining sites, flotation plant, and waste dumps has increased Rn concentrations in production areas and surrounding residential areas. In the operating room of the deputy manager of the flotation plant, the concentration of Rn in the house was 247Bq/m^3, and 156Bq/m^3 outside the house. In some houses, the concentration of Rn in the house was measured from 237-278Bq/m^3 and outside the house was measured from 105 - 208Bq/m^3.

The results of calculating effective dose in households surrounding the mine area and working area show that the majority of households and working areas of employees in Sin Quyen copper factory must receive a dose of over 10mSv/year. As recommended by the ICRP, this is the dose level for which intervention actions should be considered.

4.2.2.2. Evaluation of the effect of dose variation

The production area (A group staff) has a dose variation of 2.48 mSv/year, nearly 10 times lower than the permitted standard. Therefore, the production area (the field, the flotation plant) is currently radioactively unsafe. Meanwhile, the situation in scattered residential areas located in high and open areas of the terrain, the new variable dose is at 0.49 mSv/year, lower than the allowable safe level. Meanwhile, in resettled residential areas, the average dose calculation results are up to 6.25 mSv/year, accounting for 75% of the total effective dose. The value of effective dose variation for the population in the resettlement area reached values from 3.4 to 8.04 mSv/year, an average of 5.63 mSv/year (Table 4.6), approximately 5.5 times higher than that of the population in the resettlement area approved safety standards.

4.3. Proposing solutions to prevent harmful effects of radioactive environment caused by mining and processing of radioactive minerals

4.3.1. Total prevention solution

- Transfer investigation results to local authorities at all levels.
- Propaganda and raise people's knowledge about radiation safety.
- Carrying out environmental impact assessment on socio-economic activities.
- Residential planning:
- Use of water source:
- Agricultural production:
- Problems and mining and processing:

4.3.2. Specific preventive solutions at Sin Quyen copper mine, Lao Cai

- To reduce the annual committed effective dose, it is necessary to use measures to reduce the concentration of radioactive gas in the house in the immediate future, such as using ventilation and opening windows.
- Should not use groundwater, surface water in the area and the government needs to provide clean water.
- Spray water to reduce dust during copper ore mining and processing.

CONCLUSIONS AND RECOMMENDATIONS

1. Conclusion

From the research results of the thesis, the postgraduate draws some main conclusions as follows:

1.1. The methods to study the dispersion characteristics and variation of the radioactive environment due to the mining and processing activities of bearing radioactive ores are proposed and applied. The including methods are surveying the radioactive environment, sampling and mineral, geochemical and radiological analysis of the water, soil, ore, and rock samples which helps to clarify the differences in the characteristics of the geochemical environment in mining activities (weakly alkaline environment rich in HCO_3^-) and processing (acidic sulfide-rich environment with low pH) to increase the possibility dissolve of uranium in copper ore. This is a reasonable and scientific research method.

1.2. Determining the existence form of uranium as uraninite mineral symbiotic with copper ore helps to clarify the characteristics of uranium dispersion in the water environment of Sin Quyen copper mine.

1.3. Building a model from theory to experiment to help predict the radon release from the Sin Quyen copper deposit area to the resettled residential area near the mine. The results of the calculation of radon concentrations in the resettled residential areas have a high correlation and are consistent with the actual data of accumulated radon measurement by detector CR-39 (correlation coefficient R=0.95).

1.4. The characteristics of radioactive dispersion in the water environment due to the mining and processing of copper ore at Sin Quyen deposit are determined as follows:

- When exploiting, the water at the field site has a high content of HCO3- anion (from 30 to 292 mg/l, average 125mg/l) and pH from 6.3 to 8.75, average 7.3, with a weak alkaline environment features that increases the solubility of uranium from the solid mineral uraninite by dozens of times.
- When processing copper ore, pyrite and other sulfide minerals are crushed and mixed with the ore to create a sulfuaric acid environment that decreased the pH from 7.3 to 2.7 (acidic environment) and strongly increases solubility uranium up hundreds of times.

1.5. Copper ore mining and processing activities cause dispersion and change in uranium content in soil, water, and radon concentrations in the air, along with changes in gamma radiation intensity, causing changes in irradiation doses mine, flotation plant and surrounding residential areas.

1.6. The results of assessment of radioactive environmental effects caused by mining and processing of copper ore at Sin Quyen deposit help radioactive activity determination at the mining site, the flotation plant area, landfilled with uranium content exceeding the allowable safety standards for drinking water. The resettlement site, more than 1 km from the mine site, is located in low-lying terrain, where the radon concentration inside and outside the house exceeds the allowable safety standards. The increase in irradiation doses in the resettlement area is 5.63 mSv/year, exceeding 5.63 times the allowable safe level for residents, in which the radiation dose component through inhalation reaches 75% total doses value.

2. Recommendations

2.1. The methods for studying dispersion characteristics and assessing environmental effects due to mining and processing activities proposed in the thesis are reliable and suitable with the technical and economic conditions of our country. At present, it should be applied when there are activities of exploration, extraction, mining and processing of radioactive minerals.

2.2. The results of the assessment of radioactive environmental impacts due to the mining and processing activities of copper ore at Sin Quyen deposit can be obtained transferred to local authorities to serve the socio-economic development planning and protect public health.

2.3. The application of the M5P algorithm to study the characteristics of radon release in the thesis is only for method construction and initial testing because the new research data was conducted at only 21 houses in the mine area, however, the result has shown the effectiveness and applicability of algorithms in solving problems of radon release from the mine area to the surrounding environment. Therefore, the postgraduate proposed to continue to have further studies on the applicability of algorithms to calculate and predict the radon release from the mine area to the surrounding environment.

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