http://www.journalssystem.com/ppmp

# Characterization and concentration of U, Nb, Pb, Ag, and Fe from the Huayangchuan polymetallic ore

# Hongwei Cheng, Zihu Lv, Dongyin Wu, Dengkui Zhao

<sup>1</sup> Zhengzhou Institute of Multipurpose Utilization of Mineral Resources, CAGS, Zhengzhou, 450006, Henan, China

<sup>2</sup> Key Laboratory for Polymetallic Ores' Evaluation and Utilization, MNR, Zhengzhou, 450006, Henan, China

<sup>3</sup>Northwest China Center for Geosience Innovation, Xi'an, 710054, Shanxi, China

Corresponding author: lvzihu1980@163.com (Zihu Lv)

**Abstract:** The separation and recovery of valuable metals from the Huayangchuan uranium polymetallic ore was investigated based on mineralogical research and mineral processing experiments. The most promising valuable elements in the ore, according to mineralogical studies, are U, Nb, and Pb. 95.75% of U and 93.00% of Nb are found in betafite, 46.85% of Pb is present in galena, 52.01% of Pb occurs in cerussite, and associated Ag exists primarily in galena. The beneficiation process involving gravity concentration, magnetic separation, and flotation was determined based on the mineralogical characteristics of the ore. The U-Nb concentrate with U grade of 3578 ppm and recovery of 83.18 %, Nb<sub>2</sub>O<sub>5</sub> grade of 4391 ppm and recovery of 74.55 % can be obtained, and five elements including U, Nb, Pb, Ag, and Fe are recovered. Compared with the previous beneficiation experimental process, the flowsheet is greatly simplified and the beneficiation recovery efficiency has been improved. The results of this investigation can help to address the gaps in the processing of low-grade uranium deposits with similar mineralogical properties.

*Keywords:* betafite, dense medium cyclone, Huayangchuan, uranium polymetallic ore, separation and recovery

# 1. Introduction

Uranium (U) is a crucial component of the world's energy supply and a fundamental raw element for the growth of the nuclear industry, which is primarily used to produce nuclear fuel and weapons (Songqing et al., 2019; Yang et al., 2021). Niobium (Nb) is an indispensable metal material for strategic emerging industries and has excellent properties such as high-temperature resistance, corrosion resistance, wear resistance, superconductivity, good mechanical properties, and so on, which is widely used in steel, high-temperature alloy, biomedical material, nuclear energy, aeronautics & astronautics (Linnen et al., 2014; Schulz, 2017). Uranium and niobium are of great significance to national security and the development of emerging industries. Their demand and consumption have been high and rising continuously, and both metals have been classified as crucial strategic minerals or critical minerals by developed economies such as the United States, Europe, and Japan (Gislev et al., 2018; Gulley et al., 2018).

U is only found at around 2.7 ppm and Nb is only found at about 24 ppm in the earth's crust (Jiang et al., 2023; Mwalongo et al., 2023). As of 2022, there are 4.688 million tons of reasonably assured resources (RAR) of uranium with a global cost of less than USD 260 per kilogram and 3.229 million tons of inferred resources (N.E. Agency, 2023). With 65% of the world's total resources, the top five uranium-producing nations are Australia, Canada, Kazakhstan, Niger, and Namibia. Australia's RAR uranium resources, which total 1.318 million tons and make up 28.11% of the world's resources, are among them. China's RAR uranium resources are 111,100 tons, accounting for 2.37% of the global uranium resources, ranking 12th in the world. The total amount of uranium resources in the world has increased significantly, but there are few uranium resources with high grade and low mining costs, accounting

for less than 20 % of the world's total (N.E. Agency, 2023). Therefore, it is necessary to strengthen the development and utilization of low-grade uranium resources.

In order to economically develop low-grade uranium resources, first of all, pre-concentration methods should be used to increase the content of uranium minerals and reduce subsequent metallurgical costs (Bhargava et al., 2015). Secondly, it is necessary to recover as many associated valuable components as possible to maximize the economic value of resources. Studies have shown that when the grade of  $U_3O_8$  in the ore is increased from 0.1 % to 0.2 %, the direct processing cost per pound of  $U_3O_8$  can be saved by nearly half (Edwards and Oliver, 2000). Therefore, the use of mineral processing methods to improve the grade of uranium ore before hydrometallurgy has attracted the attention of many countries. At present, the beneficiation methods of different types of uranium ores mainly include radioactive separation and conventional beneficiation (Chen, 1989). Radioactive separation is mainly used for discarding coarse-grained tailings of hard rock-type uranium ore with uneven mineralization and low grade, which is currently used in many countries (Kidd and Wyatt, 1982; Lunt and El-Ansary, 2007). Conventional mineral processing methods mainly include gravity separation, magnetic separation, and flotation.

Uranium deposits can be classified into 15 types according to their geological characteristics, many of which can be further divided into several subtypes (Dahlkamp, 2013). And most types can be found in China. The most important types of uranium deposits in China are sandstone type (43%), granite type (22.9%), volcanic rock type (17.6%), and carbonate rock type (8.7%), accounting for more than 92.2% of China's uranium reserves (Xu et al., 2021). Carbonatite is an economically important rock, often associated with deposits of light rare earth elements (LREE) and rare metals, and nearly 20 % of carbonatites show the geochemical enrichment characteristics of rare metals and LREE. For example, the Baiyunbo deposit, the world's largest rare-earth, and second-largest niobium deposit, as well as one of China's large iron and thorium deposits, is associated with carbonated rocks (Xu et al., 2020).

The Huayangchuan carbonate-type uranium-niobium deposit is the first super-large hard rock-type uranium polymetallic deposit discovered in China, associated with niobium, lead and other metal elements (Bin-Yue et al., 2018; Cai et al., 2020). The uranium content in the ore is about 160 ~ 300 ppm (Shoujing, 2020). Several studies have been conducted on the Huayangchuan carbonate-type uraniumniobium deposit, amongst which are the works of the following. Liu et al. (2021b) used the combined process of spiral, shaking table and MD3 Knelson centrifugal concentrator to recover valuable elements uranium, niobium, lead, and iron. The recovery rates of uranium, niobium, lead, and iron in the concentrate were 81.29 %, 82.83 %, 81.33 %, and 71.89 %, respectively, and the tailing yield was 82.28 %, but the uranium grade was low and the enrichment ratio was only 4.59. Further methods utilized by Liu et al. (2021a) included flotation to recover lead from gravity concentrate, magnetic separation to recover magnetite from lead tailings, and flotation to recover uranium-niobium minerals from iron tailings using benzohydroxamic acid (C7H7NO2) as a collector. Although good beneficiation indexes were obtained, qualified uranium-niobium concentrate was obtained by multi-stage gravity separation and flotation, the process was complicated. The enrichment ratio of uranium in the combined process of gravity separation is not high, and the amount of tailings thrown by gravity separation is not enough. The high yield of gravity separation concentrate leads to a large amount of flotation feed and a large consumption of reagents. Additionally, benzohydroxamic acid is expensive, unsustainable, and requires extensive wastewater treatment (Han et al., 2021; Meng et al., 2023). Huang (2006) used the jigging-based gravity separation process to discard the tailings, and 80.58 % of the tailings were discarded to obtain a rough concentrate containing uranium, niobium, lead, and sulfur. Then the magnetic separation and flotation process were used successfully for separation, and concentration of the lead, iron, and sulfur. The obtained uranium-niobium concentrate contains U 1840 ppm, Nb<sub>2</sub>O<sub>5</sub> 2780 ppm, lead concentrate contains Pb 63.14%, iron concentrate contains TFe 72.19%, and sulfur concentrate contains S 51.20% (Huang, 2006). However, there are also some problems, such as complex processes, insufficient tailings discarding in gravity separation, low grade of uranium-niobium concentrate, and high cost of direct metallurgical leaching. Li et al. (2019) used a flotation process to increase the grade of U to 377 ppm niobium titanium uranium ore to 5465 ppm and obtained promising beneficiation results. However, the flotation process requires large amount of reagents, and the collector benzohydroxamic acid raises environmental concerns (Dong et al., 2022).

The specific purpose of this study is to develop a physical beneficiation process for the separation and recovery of valuable metals from the Huayangchuan uranium polymetallic ore, reduce subsequent metallurgical costs, and enhance the value of uranium resources. Firstly, the mineralogical characteristics of the ore were studied by chemical analysis and mineral liberation analysis (MLA). Then the separation and recovery process of U, Nb, Pb, Ag, and Fe was developed according to the mineralogical characteristics of ore, and the recovery index of U, Nb, Pb, Ag, and Fe was examined. The results of this investigation can help to address the gaps in the processing of low-grade uranium deposits with similar mineralogical properties.

## 2. Materials and methods

#### 2.1. Materials

For this study, a 2-ton representative ore sample from the Huayangchuan deposit was taken. For mineralogical and beneficiation experiments, it was initially crushed to a top size of 3 mm using a combination of jaw crusher, sieving, and roll crusher.

The chemical composition of the ore is shown in Table 1. The X-ray diffraction pattern of the ore is displayed in Fig. 1. It can be seen from Table 1 that the contents of U, Nb<sub>2</sub>O<sub>5</sub>, Pb, Ag, and Fe are 313 ppm, 405 ppm, 0.66%, 3.98 ppm, and 2.55%, respectively, which indicates that the deposit is a low-grade U-Nb polymetallic deposit. Apart from them, the other main components are SiO<sub>2</sub> (64.26%), Al<sub>2</sub>O<sub>3</sub> (8.92%), CaO (5.71%), K<sub>2</sub>O (5.09%), BaO (1.92%) and Na<sub>2</sub>O (1.25%).

Table 1. Chemical analysis of the ore sample

Component	U*	Nb <sub>2</sub> O <sub>5</sub> *	Pb	S	TFe	TiO <sub>2</sub>	CaO	MgO	K <sub>2</sub> O	Na <sub>2</sub> O
Content wt.%	313	405	0.66	0.58	2.55	0.22	5.71	0.71	5.09	1.25
Component	$Al_2O_3$	SiO <sub>2</sub>	Mn	$P_2O_5$	SrO	BaO	$RE_{x}O_{y}$	Ag*	LOI	/
Content wt.%	8.92	64.26	0.15	0.17	0.87	1.95	0.09	3.98	6.75	/

\* ppm



Fig. 1. X-ray diffraction patterns of the ore sample

# 2.2. Analysis and characterization

The chemical composition of the ore was ascertained using inductively coupled plasma-atomic emission spectroscopy (Intrepid II XSP, Thermo Electron, USA). The mineralogical composition and content of the ore sample were determined by methods including optical microscopy analysis (ZEISS Axioskop

40, Zeiss, Oberkochen, Germany), X-ray diffraction analysis (Rigaku D/Max –1200 X-ray diffractometer, Japan), and Mineral Liberation Analyzer (MLA 650F, FEI Company, Hillsboro, OR, USA). The mass distribution by size fraction of the main metal minerals in the ore was extracted from MLA and an optical microscope, and the results are shown in Table 3.

X-ray diffraction analysis was performed on a Rigaku SmartLab X-ray diffraction spectrometer employing a graphite-filtered Cu K $\alpha$  radiation ( $\lambda$  =1.5406 Å), operated at 40 kV and 40mA with the scanning rate of 5°/min from 2° to 70°. Electron probe analysis was conducted using EPMA-1720 (Shimadzu) with a beam current of 10 nA and an accelerating voltage of 20 kV.

# 2.3. The elemental deportment

The elemental deportment in different mineral species has been obtained by MLA, electron probe microanalysis, and theoretical calculation of ore composition. Firstly, The mineral composition and content were obtained by MLA. Then the main elements contents in different minerals were acquired by electron probe microanalysis or Theoretical chemical composition of minerals. The U contents in betafite and plumbobetafite were obtained by electron probe analysis, and the U content of uraninite was acquired by the theoretical chemical composition of minerals. The Pb content in galena was obtained by the theoretical chemical composition of the mineral, and the Pb contents in other minerals were acquired according to the results of electron probe analysis. Finally, the distribution of the main elements in minerals is obtained by equilibrium calculation.

The sample preparation process for MLA as follows. First, the sample was ground to a certain fineness and divided into + 0.074mm, 0.045-0.074mm, and -0.045mm fractions using standard sieves, and the yield of each fraction was calculated. Then 2.0 g dried mineral powder of each particle size was respectively mixed with epoxy resin and curing agent. To obtain the MLA resin sample, they were placed to a plastic mold for mixing, heating, 24 hour placement, and curing. The polisher's polishing strength was assumed to be 30 N, the rotating speed was 200 r/min, and the abrasive paper grit was 300, 600, 800, and 1200 grit. Polishing Solution was applied sequentially to the sample surface to polish it further. The polished samples were cleaned, dried and covered with carbon and analyzed by MLA. Finally, the MLA analysis results of the whole sample were integrated and calculated according to the yield of the three size fractions.

The elemental deportment (P) of a given element in mineral phase *i* can be calculated using equation Eq. (1) and Eq. (2).

$$P_i = \frac{W_i A_i}{A_0} \times 100\% \tag{1}$$

$$A_0 = \frac{\sum W_i A_i}{\sum W_i} \tag{2}$$

where  $W_i$  refers to the content of mineral *i* in the raw ore sample;  $A_i$  and  $A_0$  represent the elements contents in different minerals and raw ore, respectively (wt%).

#### 2.4. Ore pre-concentration

Analysis indicates that the minerals of interest including betafite  $(3.75-4.82 \text{ t}\cdot\text{m}^{-3})$ , plumbobetafite  $(4.64 \text{ t}\cdot\text{m}^{-3})$ , thorite  $(4.4-5.4 \text{ t}\cdot\text{m}^{-3})$ , galena  $(7.4-7.6 \text{ t}\cdot\text{m}^{-3})$ , cerussite  $(6.55 \text{ t}\cdot\text{m}^{-3})$  and magnetite  $(4.9-5.2 \text{ t}\cdot\text{m}^{-3})$ , all have a higher density than gangue minerals such as quartz  $(2.65 \text{ t}\cdot\text{m}^{-3})$ , orthoclase  $(2.55-2.63 \text{ t}\cdot\text{m}^{-3})$ , albite  $(2.6-2.63 \text{ t}\cdot\text{m}^{-3})$  and calcite  $(2.71-2.94 \text{ t}\cdot\text{m}^{-3})$ . The concentration criterion [CC = (SG of interest mineral – SG of fluid )/ (SG of gangue – SG of fluid )] for minerals of interest that are processed by gravity separation is given in Table 2. It shows that the CC values of several minerals are all above 2, indicating that these minerals are relatively easy to separate by gravity techniques, and the separation lower limit of particle size is 0.15 mm (Wills and Finch, 2016).

Dense medium cyclone (DMC) has been demonstrated to be a successful preconcentration method for treating lead-zinc ore, copper ore, tungsten ore, and other minerals (Chen et al., 2014; Umucu and Deniz, 2012). The low grade U-Nb polymetallic ore in this investigation was pretreated using a gravity-fed two-product DMC (WTMC 250/175), and the optimum parameters were established in the prior study (Lv et al., 2022). Previous study has shown that 0.3-3 mm is the optimal feed particle size for the separation performance of DMC on Huayangchuan uranium ore. Therefore, in this study, the ore

samples were first crushed to a size of 3 mm, and then they were separated into two size fractions, 0.3-3 mm, and 0-0.3 mm, by screening. Second, DMC preconcentration tests were conducted on the size fraction of 3-0.3 mm, while gravity separation (spiral and shaking table) was used to pretreat the other fraction (Cao et al., 2021; Habinshuti et al., 2021). Fig. 2 illustrates the separation process schematic flow chart.

Table 2. Concentration criterion (CC) for the minerals of interest considering a gangue SG of 2.65 in water

Mineral	SG	СС
Betafite	4.30	2.0
Plumbobetafite	4.64	2.2
Thorite	4.90	2.4
Galena	7.50	3.9
Cerussite	6.55	3.4
Magnetite	5.05	2.5



Fig. 2. Schematic flowsheet for ore pre-concentrate

## 2.5. Separation of magnetite

Magnetic separation is a very efficient way to recover magnetite since it is a strongly magnetic mineral with evident magnetic differences from other minerals in the ore (Arol and Aydogan, 2004). Tests for magnetic separation were conducted with a grinding fineness of -0.074 mm accounting for 65% and a magnetic field intensity of 0.1 Tesla. The flow schedule for recovering magnetite is displayed in Fig. 3.

# 2.6. Separation of Pb and U-Nb

The primary scientific issue after recovering magnetite from preconcentrate is to resolve the Pb and U-Nb separation and raise the grade of U-Nb concentration. Galena is a sulfide mineral with high levels of hydrophobicity; as a result, it floats better and is less likely to get wet by water. However, cerussite is a strongly hydrophilic lead-oxidized mineral. And, the surface is easy to be moist with strong hydration, and has poor floating performance. Whereas, before flotation, the vulcanizing agent can be



Fe concentrate

Fig. 3. The flowsheet for the recovery of magnetite

added for sulfurizing treatment to make Na<sub>2</sub>S or NaHS react with the surface of cerussite as follows (Hu et al., 2020; Liu et al., 2020):

$$PbCO_{3}|PbCO_{3} + Na_{2}S = PbCO_{3}|PbS + Na_{2}CO_{3}$$
(3)

$$PbCO_3 | PbCO_3 + NaHS = PbCO_3 | PbS + NaHCO_3$$
(4)

Following sulfurizing treatment, a thin layer of lead sulfide forms on the mineral's surface. The cerussite can then be separated by flotation with the addition of a collecting agent (xanthate). Bench-scale flotation experiments is performed using the flowsheet shown in Fig. 4 to recover lead minerals in order to achieve separation and enrichment with U-Nb minerals.

The flotation experiments were operated by using an XFD mechanical stirring flotation machine which has 1500, 1000, and 500 mL flotation cells (Changchun Prospecting Machinery, China). First, 500 g feed and 250 ml water were introduced to the XMB-70 rod mill, which was then ground to a fineness of -0.074 mm, accounting for 61.7% of the total. The pulp was then transferred to a 1500ml flotation cell and adjusted to 33% by adding approximately 800ml of water and stirring at 1800 rpm for 2 minutes. Sodium carbonate at 3000 g/t was used to adjust pH to 9. The sodium silicate (600 g/t) was then added and agitated for 3 minutes. Finally, ammonium dibutyl dithiophosphate and ethyl xanthate were added as collectors, and flotation was performed after stirring. Each reagent is diluted to a certain concentration before being supplied to the flotation cell. Several concentrate collections were collected consecutively, with the flotation cell of 750 mL chosen for the first concentration and the flotation cell of 500 mL chosen for the remaining concentration. The air flowrate of flotation is 0.4-0.6m<sup>3</sup>/h.

#### 3. Results and discussion

#### 3.1. Mineralogical Characteristic of raw ore

The mineral composition and content of the samples were analyzed by MLA, and the results are shown in Table 3. The results show that betafite (0.18 wt.%), galena (0.34 wt.%), and cerussite (0.51 wt.%) are the principal valuable minerals in the ore, along with a trace amount of rare earth minerals and ferrominerals. The main gangue minerals are quartz, feldspar and calcite. Other minerals are less abundant, which makes it more challenging to use them given the current state of technology and the economy.

Table 4 provides information on the mass distribution by size fraction of the main metal minerals. The findings show that more than 90% of betafite and galena are found above 0.075 mm, while the particle size of uraninite is mainly spread between 0.038 and 0.075 mm. This shows that the valuable minerals are dispersed throughout the ore with larger particle sizes, and preconcentrating ore and discarding a significant proportion of coarse tailings prior to hydrometallurgy is feasible, and it would be advantageous to increase the concentration of valuable minerals and lower the overall cost of ore treatment.



Fig. 4. Flotation flowsheet for separation of Pb and U-Nb

Table 3. The mineral phases identified and their relative proportion in the sample
--

Mineral	Chemical formula Content wt.%		Mineral	Chemical formula	Content wt.%
Betafite	(U,Ca)2(Nb, Ti)2O6 (OH)	0.18	Hornblende	(Ca,Na) <sub>2</sub> (Mg,Fe,Al) <sub>5</sub> (Al,Si) <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub>	2.50
Plumbobetafite	(Pb,U,Ca)(Ti,Nb) <sub>2</sub> O <sub>6</sub> (OH,F)	0.02	Biotite	K(Mg,Fe) <sub>3</sub> AlSi <sub>3</sub> O <sub>10</sub> (F,OH) <sub>2</sub>	1.28
Thorite	$Th(SiO_4)$	0.01	Quartz	SiO <sub>2</sub>	36.49
Uraninite	UO <sub>2</sub>	trace	Orthoclase	KAlSi <sub>3</sub> O <sub>8</sub>	27.82
Fergusonite	YNbO <sub>4</sub>	trace	Albite	NaAlSi <sub>3</sub> O <sub>8</sub>	10.72
Galena	PbS	0.34	Calcite	CaCO <sub>3</sub>	8.49
Cerussite	PbCO <sub>3</sub>	0.51	Muscovite	KAl <sub>2</sub> (Si <sub>3</sub> Al)O <sub>10</sub> (OH,F) <sub>2</sub>	0.67
Celestite	$SrSO_4$	1.67	Apatite	Ca <sub>5</sub> (PO <sub>4</sub> ) <sub>3</sub> (F,Cl,OH)	0.43
Orthite	(Ce,Ca) <sub>2</sub> (Fe,Al) <sub>3</sub> (Si <sub>2</sub> O <sub>7</sub> ) (SiO <sub>4</sub> )O(OH)	0.35	Chlorite	(Mg,Fe <sup>2+</sup> ) <sub>5</sub> Al(Si <sub>3</sub> Al)O <sub>10</sub> (OH) <sub>8</sub>	0.84
Monazite	(Ce,La,Nd,Th)PO <sub>4</sub>	0.02	Titanite	CaTiSiO <sub>5</sub>	0.61
Barite	$BaSO_4$	2.52	Zircon	ZrSiO <sub>4</sub>	0.03
Strontianite	SrCO <sub>3</sub>	0.49	Ilmenite	FeTiO <sub>3</sub>	0.01
Magnetite	Fe <sub>3</sub> O <sub>4</sub>	2.66	Garnet*	$X_{3}Y_{2}(SiO_{4})_{3}$	0.18
Limonite	FeO(OH) nH <sub>2</sub> O	0.70	Plagioclase	(Na,Ca)Al (Si,Al)Si <sub>2</sub> O <sub>8</sub>	0.09
Pyrite	FeS <sub>2</sub>	0.33	Others	/	0.25

\* X represents Ca2+, Fe2+, Mn2+, or Mg2+; Y represents Al3+, Cr3+, or Fe3+

Size Fraction (mm)	betafite	plumbobetafite	thorite	uraninite	galena	cerussite
0.5~1	20.02	0.00	0.00	0.00	23.33	0.00
0.3~0.5	21.67	0.00	0.00	0.00	22.56	13.94
0.15~0.3	35.07	0.00	0.00	0.00	30.92	26.32
0.075~0.15	16.42	0.00	11.24	0.00	18.82	18.09
0.038~0.075	2.59	10.63	25.95	60.79	2.50	11.15
0.02~0.038	1.80	11.06	9.17	4.52	0.92	10.04
0.01~0.02	1.41	20.62	18.09	12.41	0.40	9.42
< 0.01	1.02	57.69	35.55	22.28	0.55	11.04

Table 4. The mass distribution by size fraction of main minerals in the sample

The only valuable elements in the ore are uranium, niobium, lead, and silver. Using MLA, electron microprobe, and theoretical calculations of ore composition, we obtained the state and distribution of these elements among different minerals, and the results are shown in Table 5 and Table 6. It indicates that 95.75% of the uranium and 93.00% of the niobium in the ore are found in betafite, and the rest of the uranium is found in minerals such as plumbobetafite and uraninite; 46.85% of the lead in the ore occur in galena, and 52.01% of the lead occur in cerussite, but the latter has a high oxidation rate, which makes it difficult to recycle the lead. There is no independent mineral of silver in the ore. The analysis of galena in the ore shows that the silver content in the single mineral of galena reaches 630 ppm indicating that the silver in the ore mainly occurs in galena, which can be recovered with galena.

Table 5. Modal distribution of uranium and niobium

Mineral	Mineral content		U wt.%	Nb <sub>2</sub> O <sub>5</sub> wt.%		
	wt.%	Grade	Modal distribution	Grade	Modal distribution	
Betafite	0.18	21.34	95.75	27.28	93.00	
Plumbobetafite	0.02	8.53	4.25	18.47	7.00	
Uraninite	0.00	88.15	0.00	0.00	0.00	
Others	99.80	0.00	0.00	0.00	0.00	
Total	100.00	0.0401	100.00	0.0528	100.00	

Table 6. Modal distribution of lead

Mineral	Mineral content wt.%	Grade of lead in mineral wt.%	Modal distribution of lead wt.%
Galena	0.35	86.61	46.85
Cerussite	0.51	65.99	52.01
Strontianite	0.49	0.04	0.03
Betafite	0.18	0.34	0.09
Plumbobetafite	0.02	32.94	1.02
Others	98.45	0.00	0.00
Total	100.00	0.65	100.00

# 3.1.1. Characteristics of betafite

The grain size of betafite in the sample is coarse, generally greater than 0.075 mm, mainly distributed between 0.15 - 0.5 mm, and below 1 mm. The betafite in the ore is mostly straight contact with other minerals, which is conducive to its liberation. The dissemination characteristics and associated relationship of betafite are shown in Fig. 5 and Table 7, respectively. The results show that betafite is mainly associated with apatite, orthoclase, albite, calcite, and other minerals in addition to plumbobetafite.



c. Semi-automorphic granular betafite symbiotic with plumbobetafite



Fig. 5. BSE images of betafite grains showing some associations with other valuable and gangue minerals

## 3.1.2. Characteristics of pb-bearing minerals

Galena has coarse grains and is frequently found in quartz veins that also contain calcite. The majority of the lead minerals are in direct contact with the gangue minerals, but some of them are plainly oxidized, and cerussite has largely taken the place of quartz at the particle edges. Cerussite, which is the byproduct of the oxidation of the galena, is dispersed unevenly around the margin of the galena and has coarse, mainly patchy grain sizes. Additionally, some secondary cerussite is spread in the fracture in the form of threads and veins, which is a byproduct of primary cerussite weathering and leaching. The majority of the cerussite and gangue minerals share straight contact, which promotes liberation. Many cerussite monomers have been released from the -2 mm sample or have joined galena. Fig. 6 depicts the dispersal traits of pb-bearing minerals. The findings reveal that in addition to its association with cerussite, calcite, and other minerals are the principal associations of galena. Galena, calcite, and barite are the minerals that cerussite is most frequently found with.

# 3.1.3. Characteristics of magnetite

Under a reflective microscope, the magnetite appears to be primarily light gray-gray black, with a graywhite reflection hue, and uniformity. The majority of the magnetite in the ore is granular, coarse, and dispersed in the gneiss and other nearby rocks. Fig. 7 displays BSE images of magnetite grains. Statistical analysis is done to determine how 1magnetite and the other minerals in the ore relate to one another. According to the statistical findings, quartz, orthoclase, and other minerals are magnetite's principal mineral partners.

Mineral	Binary particle wt.%	Ternary particle wt.%
Plumbobetafite	18.95	0.49
Uraninite	0.02	0.07
Cerussite	0	0.7
Orthite	1.51	3.69
Monazite	0	0.01
Barite	0.25	0.02
Magnetite	0	0.24
Calcite	0.17	0.09
Albite	0.86	0.1
Orthoclase	2.26	1.84
Quartz	1.29	1.23
Hornblende	0	0.52
Muscovite	0.02	0.00
Apatite	10.15	1.22
Sphene	0.57	0.27
Chlorite	0.04	0.00

Table 7. Mineralogical associations of betafite



a. Coarse-grained galena



b. Galena and calcite symbiosis





HV mag WD 20.00 kV 400 x 12.9 mm

88

d. Filamentous cerussite

Fig. 6. BSE images of pb-bearing minerals grains showing some associations with other valuable and gangue minerals



a. Coarse-grained magnetite

b. Disseminated magnetite

Fig. 7. Polished section images of pb-bearing minerals grains showing some associations with other valuable and gangue minerals

# 3.2. Preconcentration of U-Nb, Pb, Ag and Fe

The results of the ore pre-concentration are presented in Table 8. DMC Conc. contains4141 ppm U, 4957 ppm Nb<sub>2</sub>O<sub>5</sub>, 10.09% Pb, 77.48 ppm Ag, and 22.3% Fe with the recovery of 61.35%, 53.63%, 66.16%, 67.85%, and 40.14% respectively. The enrichment ratios of U, Nb<sub>2</sub>O<sub>5</sub>, Pb, Ag, and Fe were 13.36, 11.66, 14.41, 14.76, and 8.75 respectively, and the tailings discarding ratio reached 93.75%. The results certified that DMC has good preconcentration and separation performance for the low grade ores. While, the mass yield of fine concentrates was 4.37%, and the grades of U, Nb<sub>2</sub>O<sub>5</sub>, Pb, Ag, and Fe were 1611 ppm, 2115 ppm, 1.45%, 21.22 ppm, and 5.05% with the recoveries of 22.66%, 21.74%, 9.03%, 17.65%, and 8.64% respectively. The yield of total concentrates was 8.97%, the contents of U, Nb<sub>2</sub>O<sub>5</sub>, Pb, Ag, and Fe were 2908 ppm, 3573 ppm, 5.88%, 50.07 ppm, and 13.9%, and their recoveries were 84.01%, 75.37%, 75.20%, 85.50%, and 48.88% respectively. Ore preconcentration rejects 91.03% of gangue and shows a good separation and enrichment effect in the experiments. U, Nb, Pb, and Ag are mainly enriched in gravity concentrate.

# 3.3. Recovery of magnetite

Ore preconcentration discards a large amount of gangue minerals, which saves considerable production costs for subsequent processes. The mineral composition of pre-concentrate and the magnetic susceptibility of minerals are displayed in Table 9. It indicates that the main minerals are barite (42.15%), cerussite (13.44%), galena (9.37%), quartz (4.86%), strontianite (4.44%), orthoclase (4.27%), calcite (3.47%) followed by pyrite (2.94%), betafite (2.61%), magnetite (2.58%), hornblende (2.14%), albite (1.99%), limonite (1.12%), and other minerals are less than 1%.

Magnetite has a high magnetic susceptibility of 92000×10-9 m<sup>3</sup>/kg, classifying it as a strong magnetic mineral. Low-intensity magnetic separation is the most economical and effective separation method, and the results of magnetic separation tests are listed in Table 10. It can be seen from Table 10 that the yield of magnetite concentrates obtained using low-intensity magnetic separation is 1.13%, containing 68.3% Fe, the recovery rate is 30.16%, and the enrichment ratio of Fe is as high as 27. Moreover, the separation of magnetite from pre-concentration concentrate is beneficial to increase the grade of U and Nb2O5. After magnetite recovery, the grade of U and Nb2O5 increased from 2908 ppm and 3573 ppm to 3318 ppm and 4076 ppm, respectively, and their recovery rates only lost 0.2 % and 0.17 %.

# 3.4. Separation and recovery of Pb and U-Nb

Table 11 presents the outcomes of flotation to separate Pb and U-Nb minerals. As can be observed, the Pb concentrates had a Pb content of 67.21% and an Ag content of 581.7 ppm, with recovery rates of

59.87% and 69.21%, respectively. The yield of the U-Nb concentrates is 7.22%, and their grades are 3578 ppm of U and 4391 ppm of Nb<sub>2</sub>O<sub>5</sub>, with recovery rates of 83.18% and 74.55%, respectively.

Table 8. Results of ore preconcentration by DMC and sprial-shaking table

Products	Yield	ield Grade ppm						Recovery wt.%					
	<b>wt.%</b>	U	$Nb_2O_5$	Pb*	Ag	TFe*	U	$Nb_2O_5$	Pb	Ag	TFe		
DMC Conc.	4.60	4141	4957	10.09	77.48	22.3	61.35	53.63	66.16	67.85	40.14		
DMC Tailings	69.05	38	88	0.09	0.74	1.25	8.52	14.26	8.76	9.72	33.78		
Shaking Conc.	4.37	1611	2115	1.45	21.22	5.05	22.66	21.74	9.03	17.65	8.64		
Fine tailings	5.81	46	88	0.24	1.20	1.80	0.86	1.21	1.99	1.33	4.09		
Fine slime	16.17	127	241	0.61	1.12	2.11	6.61	9.16	14.06	3.45	13.35		
Feed	100.00	310	425	0.70	5.25	2.55	100.00	100.00	100.00	100.00	100.00		

(\* wt.%)

Table 9. The mineral composition of preconcentrate and the magnetic susceptibility of minerals(Changmiao et al.,2020; Rosenblum and Brownfield, 2000)

Mineral	Content wt.%	Magnetic susceptibility( 10 <sup>-9</sup> m³/kg)
Barite	42.15	0.3
Cerussite	13.44	0.27
Galena	9.37	0.62
Quartz	4.86	0.5
Strontianite	4.44	0.14
Orthoclase	4.27	0.33
Calcite	3.47	0.37
Pyrite	2.94	26.98
Betafite	2.61	0.94
Magnetite	2.58	92000
Hornblende	2.14	25.54
Albite	11.99	62.8
Limonite	1.12	33.10

Table 10. Results of magnetite recovery by magnetic separation

Products Yield wt.%	Yield	Yield Grade ppm							Recovery wt.%					
	U	$Nb_2O_5$	Pb*	Ag	TFe*	U	$Nb_2O_5$	Pb	Ag	TFe				
Fe concentrate	1.13	55	64	0.45	3.14	68.30	0.20	0.17	0.73	0.73	30.16			
Fe tailing	7.84	3318	4076	6.66	56.77	6.08	83.81	75.20	74.47	84.77	18.72			
Feed	8.97	2908	3573	5.88	50.07	13.9	84.01	75.37	75.20	85.50	48.88			

\* wt.%.

Table 11. Results of flotation separation of Pb and U-Nb

Droducto	Yield		Grade ppm					Recovery wt.%				
Products	<b>wt.%</b>	U	$Nb_2O_5$	Pb*	Ag	TFe*	U	$Nb_2O_5$	Pb	Ag	TFe	
U-Nb concentrate	7.22	3578	4391	1.42	11.32	6.04	83.18	74.55	14.60	15.56	17.10	
Pb concentrate	0.62	309	443	67.21	581.7	6.59	0.62	0.65	59.87	69.21	1.62	
Feed	7.84	3318	4076	6.66	56.77	6.08	83.81	75.20	74.47	84.77	18.72	

\* wt.%

#### 3.5. Global results

The final process flow of separation and recovery of U, Nb, Pb, Ag, and Fe from Huayangchuan uranium polymetallic ore is shown in Fig. 8. The beneficiation process involving gravity concentration, magnetic separation, and flotation is adopted. The grade of U-Nb concentrate is U 3578 ppm and Nb<sub>2</sub>O<sub>5</sub> 4391 ppm, and the recovery rate is U 83.18 % and Nb<sub>2</sub>O<sub>5</sub> 74.55 %. The grade of Pb concentrate is Pb 67.21 %, Ag 581.7 ppm, and the recovery is Pb 59.87 %, Ag 69.21 %. The Fe concentrate with a TFe grade of 68.30 % and recovery of 30.16 % was obtained. The process can greatly reduce the grinding capacity and realize the direct obtain of U-Nb concentrate by one-step gravity separation. And five elements including U, Nb, Pb, Ag, and Fe were successfully recovered from Huayangchuan uranium polymetallic ore. The procedure is significantly more straightforward and the beneficiation recovery efficiency is higher than it was with the prior experimental beneficiation process (Huang, 2006; Liu et al., 2021a; Liu et al., 2021b).



Fig. 8. The final process flow of separation and recovery of U, Nb, Pb, Ag and Fe from Huayangchuan uranium polymetallic ore

## 4. Conclusions

The mineralogical characteristic and separation and recovery of valuable metals from the Huayangchuan uranium polymetallic ore were investigated. A physical concentration process involving gravity concentration, magnetic separation, and flotation was developed to separate and recover U, Nb, Pb, Ag, and Fe elements. The following conclusions can be drawn from this study:

The U content of the Huayangchuan uranium polymetallic ore is 313 ppm, the Nb<sub>2</sub>O<sub>5</sub> content is 405 ppm, and the Pb content is 0.66 wt%. U and Nb are mainly present in betafite, Pb is mainly present in galena and cerussite, and Ag is mainly present in galena. The dissemination size of U, Nb, and Pb minerals is coarse.

The beneficiation process involving gravity concentration, magnetic separation, and flotation is adopted, which can greatly reduce the grinding amount and realize the direct acquisition of U-Nb concentrate by one-step gravity separation of coarse crushing. The grade of U-Nb concentrate is U 3578

ppm and Nb<sub>2</sub>O<sub>5</sub> 4391 ppm, and the recovery rate is U 83.18 % and Nb<sub>2</sub>O<sub>5</sub> 74.55 %. The grade of Pb concentrate is Pb 67.21 %, Ag 581.7 ppm, and the recovery is Pb 59.87 %, Ag 69.21 %. The Fe concentrate with a TFe grade of 68.30 % and recovery of 30.16 % was obtained.

From the Huayangchuan Uranium Polymetallic ore, five elements, including U, Nb, Pb, Ag, and Fe, were successfully recovered. The procedure is significantly more straightforward and the beneficiation recovery efficiency is higher than it was with the prior experimental beneficiation process. The findings of this study can help to address the gaps in the processing of low-grade uranium deposits with similar mineralogical properties.

# Acknowledgments

The authors acknowledge the support of the Major Research Plan of the National Natural Science Foundation of China (No.91962223).

# References

N.E. Agency, Uranium 2022: Resources, Production and Demand. 2023, OECD, Paris.

- AROL, A., AYDOGAN, A., 2004. *Recovery enhancement of magnetite fines in magnetic separation*. Colloids and Surfaces A: Physicochemical and Engineering Aspects. 232(2-3), 151-154.
- BHARGAVA, S.K., RAM, R., POWNCEBY, M., GROCOTT, S., RING, B., TARDIO, J., JONES, L., 2015. A review of acid leaching of uraninite. Hydrometallurgy. 151, 10-24.
- BIN-YUE, H., RUI-QIANG, P., XING-XING, W., TAO, Z., JING, L., 2018. Geological Features of Uranium Metallization in the Huayangchuan U-Polymetallic Deposit. Acta Geologica Sichuan. 38(01), 97-100.
- CAI, J., YU, L., XU, D., GAO, C., CHEN, G., YU, D., JIAO, Q., YE, T., ZOU, S., LI, L., 2020. Multiple episodes of tectonothermal disturbances in the Huayangchuan U-Nb-Pb polymetallic deposit in the Xiaoqinling region, central China and their significances on metallogeny. Ore Geology Reviews. 127, 103755.
- CAO, M., BU, H., GAO, Y., 2021. Preconcentration of Low-Grade Ta-Nb Deposit Using Physical Separation Methods. JOM. 73, 1310-1320.
- CHANGMIAO, L., HONGWEI, C., JIAN, X., MIN, W., DONGYIN, W., ZIHU, L., DENGKUI, Z., SHOUJING, W., 2020. Study on endowment characteristics and comprehensive development and utilization technology of U-Nb-Pb polymetallic ore in Huayangchuan, Shaanxi Province. Geology in China, 1-10.
- CHEN, J., CHU, K., ZOU, R., YU, A., VINCE, A., BARNETT, G., BARNETT, P., 2014. *How to optimize design and operation of dense medium cyclones in coal preparation*. Minerals Engineering. 62, 55-65.
- CHEN, X., 1989. *Development and application of radiometric sorting to uranium ore*. Uranium Mining and Metallurgy. 8(2), 1-6.
- DAHLKAMP, F.J., Uranium ore deposits. 2013, Springer Science & Business Media.
- DONG, B., WANG, P., LI, Z., TU, W., TAN, Y., 2022. Degrading hazardous benzohydroxamic acid in the industrial beneficiation wastewater by dielectric barrier discharge reactor. Separation and Purification Technology. 299, 121644. EDWARDS, C., OLIVER, A., 2000. Uranium processing: a review of current methods and technology. Jom. 52, 12-20.
- GISLEV, M., GROHOL, M., MATHIEUX, F., ARDENTE, F., BOBBA, S., NUSS, P., 2018. *Report on critical raw materials and the circular economy*. European Commission: Brussels, Belgium.
- GULLEY, A.L., NASSAR, N.T., XUN, S., 2018. *China, the United States, and competition for resources that enable emerging technologies*. Proceedings of the national academy of sciences. 115(16), 4111-4115.
- HABINSHUTI, J.B., MUNGANYINKA, J.P., ADETUNJI, A.R., MISHRA, B., OFORI-SARPONG, G., KOMADJA, G.C., TANVAR, H., MUKIZA, J., ONWUALU, A.P., 2021. *Mineralogical and physical studies of low-grade tantalumtin ores from selected areas of Rwanda*. Results in Engineering. 11, 100248.
- HAN, G., DU, Y., HUANG, Y., YANG, S., WANG, W., SU, S., LIU, B., 2021. Efficient removal of hazardous benzohydroxamic acid (BHA) contaminants from the industrial beneficiation wastewaters by facile precipitation flotation process. Separation and Purification Technology. 279, 119718.
- HU, Y., WU, M., LIU, R., SUN, W., 2020. A review on the electrochemistry of galena flotation. Minerals Engineering. 150, 106272.
- HUANG, M., 2006. The Experiment of Comprehensive Utilization for a Low Grade U-Nb-Pb Ore. Conservation and Utilization of Mineral Resources(04), 34-36.
- JIANG, S.-Y., WANG, W., SU, H.-M., 2023. Super-Enrichment Mechanisms of Strategic Critical Metal Deposits: Current Understanding and Future Perspectives. Journal of Earth Science. 34(4), 1295-1298.

KIDD, D., WYATT, N., 1982. Radiometric sorting of ore.

- LI S., ZHU Y., FANG, C., HU J., CUI Q., GUIBING, Z., 2019. *Experimental Study on Recovery of Fine Mendeleevite*. Modern Mining. 35(03), 105-109+153.
- LINNEN, R., TRUEMAN, D.L., BURT, R., 2014. Tantalum and niobium. Critical metals handbook, 361-384.
- LIU, R., LIU, D., LI, J., LIU, S., LIU, Z., GAO, L., JIA, X., AO, S., 2020. Improved understanding of the sulfidization mechanism in cerussite flotation: An XPS, ToF-SIMS and FESEM investigation. Colloids and Surfaces A: Physicochemical and Engineering Aspects. 595, 124508.
- LIU Z., LI C, JIA X., LI G., QIANG L., MA J., WU C., BAOBIN, T., 2021a. Study on Comprehensive Recovery Technology of Hua Yangchuan Low Grade U Nb Pb Polymetallic Ore. Conservation and Utilization of Mineral Resources. 41(03), 132-137.
- LIU Z., LI ., LI C., QIANG L., MA J., TIAN Y., WU C., BAOBIN, T., 2021b. Study on Gravity Separation Recovery Technology of Huayangchuan Low Grade Uranium Polymetallic Ore Uranium Mining and Metallurgy. 40(04), 273-277.
- LUNT, D., BOSHOFF, P., BOYLETT, M., EL-ANSARY, Z., 2007. *Uranium extraction: the key process drivers*. Journal of the Southern African Institute of Mining and Metallurgy. 107(7), 419-426.
- LV, Z., WEI, M., ZHAO, D., WU, D., LIU, C., CHENG, H., 2022. *Preconcentration of a low-grade betafite ore by dense medium cyclone*. Physicochemical Problems of Mineral Processing. 58.
- MENG, X., JIANG, M., LIN, S., GAO, Z., HAN, H., TIAN, M., ZHANG, C., LIU, R., WU, M., BAO, H., SUN, W., 2023. Removal of residual benzohydroxamic acid-lead complex from mineral processing wastewater by metal ion combined with gangue minerals. Journal of Cleaner Production. 396, 136578.
- MWALONGO, D.A., HANEKLAUS, N.H., LISUMA, J.B., KIVEVELE, T.T., MTEI, K.M., 2023. Uranium in phosphate rocks and mineral fertilizers applied to agricultural soils in East Africa. Environmental Science and Pollution Research. 30(12), 33898-33906.
- ROSENBLUM, S., BROWNFIELD, I.K., *Magnetic susceptibilities of minerals*. 2000, US Department of the Interior, US Geological Survey.
- SCHULZ, K.J., Critical mineral resources of the United States: economic and environmental geology and prospects for future supply. 2017, Geological Survey.
- SHOUJING, W., 2020. *Study on the Occurrence of the Useful Element in Huayangchang Uranium Polymetallic Ore.* Conservation and Utilization of Mineral Resources. 40(4), 6.
- SONGQING, L., YANGGE, Z., SHUANFANG, C., JINPIN, H., QIANG, C., GUIBIN, Z., 2019. Experimental Study on Recovery of Fine Mendeleevite. Modern Mining. 35(03), 105-109.
- UMUCU, Y., DENIZ, V., 2012. A study simulation and modeling on the performance of the heavy media cyclone in coal *beneficiation*. AGH Journal of Mining and Geoengineering. 36(4), 179-185.
- WILLS, B.A., FINCH, J.A., 2016. Gravity Concentration, In Wills' Mineral Processing Technology. Elsevier, pp. 223-244.
- XU, D., CHI, G., NIE, F., FAYEK, M., HU, R., 2021. Diversity of uranium deposits in China–An introduction to the Special Issue. Ore Geology Reviews. 129, 103944.
- XUE, S., LING, M.-X., LIU, Y.-L., KANG, Q.-Q., HUANG, R.-F., ZHANG, Z.-K., SUN, W., 2020. The formation of the giant Huayangchuan U-Nb deposit associated with carbonatite in the Qingling Orogenic Belt. Ore Geology Reviews. 122, 103498.
- YANG, Y., RAM, R., MCMASTER, S.A., POWNCEBY, M.I., CHEN, M., 2021. A comparative bio-oxidative leaching study of synthetic U-bearing minerals: Implications for mobility and retention. Journal of Hazardous Materials. 403, 123914.