

Beneficiation Studies of a Rare Earth Ore from the Olserum Deposit

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Abstract

The Olserum deposit located in Sweden hosts an indicated mineral resource of 4.5 million tonnes at 0.6% total rare earth oxides (TREO), exhibiting a relatively high proportion of heavy rare earth elements (HREE). The mineralogy and beneficiation of a composite drill core sample from the Olserum deposit were studied. Monazite and xenotime were found by the mineralogical analyses to be the target minerals for beneficiation of REEs and most (96.9%) of REEs in the ore are carried by monazite (68.5%) and xenotime (28.4%). Because xenotime carries 84.7% of heavy REEs (Y, Gd, Dy) in the ore it is more valuable mineral than monazite. A beneficiation process was developed which includes grinding, wet low intensity magnetic separation (WLIMS) for removal of magnetite and REEs flotation consisting of one stage of roughing and two stages of cleaning. Selective flotation collector of REE minerals and suitable grinding size of feed material were determined by testwork. The REE concentrate and tailings were chemically and mineralogically characterized. The studies of process mineralogy showed that the REE-bearing minerals, monazite and xenotime, and apatite were successfully enriched from the concentrations of 0.6%, 0.31% and 2.6% in the feed to those of 17.0%, 8.9% and 65.0% in the concentrate with the recoveries of 79.0%, 81.3% and 71.0%, respectively.

Keywords

The Olserum Deposit, Rare Earth Minerals, Flotation

1. Introduction

Rare earth elements (REEs) normally refer to 15 lanthanides plus chemically similar yttrium (Y) and scandium (Sc). REEs typically split into two sub-groups, the light rare earth elements (LREE) which includes Sc, La, Ce, Pr, Nd, Pm, Sm

and Eu, and the heavy rare earth elements (HREE) which includes Y, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu. The Olserum deposit located in Sweden comprises lower grade placer material with higher grade apatite-magnetite veins and hosts an indicated mineral resource of 4.5 million tonnes at 0.6% total rare earth oxides (TREO), with 34% of that being heavy rare earth oxides (HREO) [1]. The mineralization was studied by Andersson *et al.* (2018) [2]. At early stage, high-temperature REEs were characterized by abundant monazite-(Ce) and xenotime-(Y) coexisting with fluorapatite and subordinate amounts of (Y, REEs, U, Fe)-(Nb, Ta) oxides. During a subsequent stage, allanite-(Ce) and ferriallanite-(Ce) were formed locally, partly resulting from the breakdown of primary monazite-(Ce). Alteration of allanite-(Ce) or ferriallanite-(Ce) to bastnäsite-(Ce) and minor synchysite-(Ce) at lower temperatures represented the latest stage of REE mineral formation.

The mineralogy and beneficiation of the ore were firstly studied by GTK Mintec in the EURARE project for a composite drill core sample. The Olserum is considerably smaller than the NorraKärr deposit. However, rare earth elements (REEs) are hosted by xenotime and monazite, both of which have well established processing pathways, and meanwhile, the Olserum has a high contribution of the high value critical REEs (dysprosium (Dy), yttrium (Y), neodymium (Nd) and terbium (Tb)), which provide a unique advantage for its exploration [3]. In this paper, flotation of the REE-bearing minerals in the Olserum ore was studied.

2. Materials and Methods

A drill core composite sample weighted 110 kg from the Olserum deposit was sent to GTK Mintec in Outokumpu, Finland. The sample was crushed to -1.5 mm and homogenized, and split to 1.5 kg subsamples. One subsample of 1.5 kg was further split into 100 g samples for chemical and mineralogical analyses.

Sodium peroxide plus sodium hydroxide digestion was used for determination of REEs by ICP-MS technique which was performed by Labtium Oy in Finland. The limits of detection (LODs) of the ICP-MS for REEs are shown in **Table 1**.

Quantitative mineralogical studies were carried out by using a mineral liberation analyzer (MLA) and an electron probe micro analyzer (EPMA).

The modal mineralogy, *i.e.* the percentages of the mineral components, the mineral liberation and the grain size distribution of REE-bearing minerals were measured using MLA (a FEI MLA Quanta 600 system) at GTK Mintec. Minerals were identified mostly based on Energy Dispersive X-Ray Spectroscopy (EDS) analyses. Using the EDS analysis the composition of a certain mineral was determined and the composition was compared to the composition of the mineral in MinIdent mineralogy database and a mineralogical book [4]. Also the EDS spectrum was compared to the spectra in the databases of MLA.

For determining the chemical compositions of REE minerals in the sample a CAMECA SX100 EPMA at the Geological Survey of Finland in Espoo, Finland was used. Analytical results were corrected using the PAP on-line correction

programme [5]. The limits of detection (LODs) of the EPMA for REEs and radioactive elements, U and Th, are shown in **Table 2**.

The subsamples of 1.5 kg with the size of -1.5 mm were ground at a laboratory rod mill as the feeds for beneficiation experiments. The relationships between grinding size and time were determined by testing. The concentration of solid for grinding was 60%wt using tap water at the room temperature (20°C - 23°C).

According to the mineralogical properties of the sample the flowsheet of magnetic separation of magnetite followed by REE-bearing minerals flotation was designed. Flotation experiments were carried out using a GTK laboratory flotation machine. The flotation cells with the volumes of 1.5, 2.5 and 4.0 liters were used. Tap water was used for conditioning and flotation at the room temperature. Wet low intensity magnetic separation (WLIMS) experiments were performed using a Sala laboratory magnetic separator (wet low intensity roller type) with the magnetic field of 0.07 Tesla.

3. Results and Discussion

3.1. Chemical and Mineralogical Analyses

Chemical analysis results are shown in **Table 3**. It is seen that the content of total REEs (TREE) in the Olserum composite ore is only 0.54%, but the rate of valuable heavy REEs (HREE) is quite high, that is, 37% of REEs in the ore are HREE. The radioactive elements, U and Th, have contents of 242 mg/kg and 81 mg/kg, respectively.

Total 28 minerals were identified by MLA. The modal mineralogy is presented in **Table 4**. It is revealed that this is a high silicate ore with the silicate concentration of 94%. In addition, a relatively high content of magnetite (2.29%) was recognized.

Four minerals were identified by MLA and EPMA to bear REEs, two phosphate minerals monazite-(Ce) and xenotime with the contents of 0.61% and 0.31%, respectively, one silicate mineral allanite with the content of 0.07%, and one niobium oxide mineral pyrochlore with the content of 0.01%.

The chemical compositions of the four REE-bearing minerals were measured by EPMA. The results (**Table 5**) show that compositions of REO in these four REE-bearing minerals are 65.4% (monazite), 53.4% (xenotime), 25.1% (allanite) and 3.3% (pyrochlore). Monazite and xenotime have high compositions of REO, especially xenotime also being high in the concentration of heavy REO including Y_2O_3 , Gd_2O_3 and Dy_2O_3 .

Based on the data of modal mineralogy and the REO compositions of REE-bearing minerals the concentrations of REEs (measured by REOs) in the ore were calculated and shown in **Table 6**. It is seen that the calculated concentrations based on the mineralogical analyses of MLA and EPMA are very close to the chemical analysis data by ICP-MS. The distributions of total REEs (measured by REOs) in the REE-bearing minerals are: 68.5% in monazite, 28.4% in xeno-

time, 3.0% in allanite and 0.1% in pyrochlore, respectively. The distributions of heavy REEs (Y, Gd and Dy) in the REE-bearing minerals are: 14.5% in monazite, 84.7% in xenotime, 0.6% in allanite and 0.2% in pyrochlore, respectively.

Table 1. The limits of detection (LODs) of the ICP-MS for REEs (mg/kg).

La	Ce	Pr	Nd	Sm	Eu	Gd	Tb
0.1	0.1	0.1	0.2	0.2	0.05	0.05	0.1
Dy	Ho	Er	Tm	Yb	Lu	Y	Sc
0.1	0.1	0.05	0.1	0.05	0.1	1	1

Table 2. The limits of detection (LODs) of the EPMA for REEs (mg/kg).

La	Ce	Pr	Nd	Sm	Gd	Dy	Y	Sc	U	Th
2016	2365	3122	4547	3571	3366	2368	1711	529	3225	2770

Table 3. Chemical analysis results (mg/kg).

La	Ce	Pr	Nd	Sm	Eu	Gd	Tb
621	1590	202	796	187	3	209	36
Dy	Ho	Er	Tm	Yb	Lu	Y	Sc
217	45	132	19	123	0	1210	0
ZrO ₂					TREE		HREE/LREE
%	U	Th				wt%	
0	242	82	0.08		5390	0.54	37/63

Table 4. Modal mineralogy of the Olserum ore (wt%).

	Silicate		93.94
Quartz	47.89	Chamosite	0.31
Albite	11.9	Illite	0.92
K_feldspar	0.54	Andalustite	2.12
Cummingtonite	0.9	Staurolite	1.05
Gedrite	3.96	Tourmaline	0.25
Biotite	19.66	Allanite ^a	0.07
Muscovite	0.29	Zircon	0.02
Clinochlore	0.65		
	Carbonate		0.10
Calcite	0.04	Siderite	0.06
	Phosphate		3.56
Apatite	2.59	Xenotime ^a	0.31
Monazite-(Ce) ^a	0.61	Wagnerite	0.05
	Other		2.34
Magnetite	2.29	Pyrochlore ^a	0.01
Ilmenite	0.04	Masuyite	0.00
Unclassified			0.06
	Total		100.00

^aREE-bearing minerals.

Table 5. The individual concentrations of REEs and U and Th in all the REE-bearing phases: allanite, monazite, xenotime and pyrochlore by EPMA (mg/kg) (measured by REOs, UO₂ and ThO₂)^a.

	Monazite	Xenotime	Allanite	Pyrochlore
UO ₂	12,100	6200		159,400
ThO ₂	13,300	n.d.		3200
La ₂ O ₃	114,000	n.d.	41,400	n.d.
Ce ₂ O ₃	311,700	n.d.	125,100	n.d.
Pr ₂ O ₃	32,400	n.d.	13,000	n.d.
Nd ₂ O ₃	124,600	n.d.	47,200	n.d.
Sm ₂ O ₃	25,100	7700	8800	n.d.
Y ₂ O ₃	20,300	410,500	8100	17,500
Gd ₂ O ₃	19,600	51,600	7000	8500
Dy ₂ O ₃	5900	63,800	n.d.	7300
Total REO	653,700	533,600	250,600	33,300
HREO	45,800	525,900	15,100	33,300

^an.d.: not detected.**Table 6.** The concentrations of REOs, UO₂ and ThO₂ in the ore (mg/kg)^a.

	Monazite	Xenotime	Allanite	Pyrochlore	Total (MLA-EPMA)	ICP-MS
UO ₂	73.7	19.3		15.9	109.0	274.5
ThO ₂	81.2	n.d.		0.3	81.5	92.2
La ₂ O ₃	695.1	n.d.	29.0	n.d.	724.1	728.3
Ce ₂ O ₃	1901.6	n.d.	87.5	n.d.	1989.1	1862.3
Pr ₂ O ₃	197.8	n.d.	9.1	n.d.	206.9	236.4
Nd ₂ O ₃	760.1	n.d.	33.0	n.d.	793.1	928.4
Sm ₂ O ₃	153.2	23.9	6.2	n.d.	183.4	216.8
Y ₂ O ₃	124.1	1272.5	5.7	1.7	1404.0	1536.6
Gd ₂ O ₃	119.5	160.0	4.9	0.8	285.2	240.9
Dy ₂ O ₃	36.1	197.9	n.d.	0.7	234.8	249.0
Total REO	3987.6	1654.5	175.4	3.3	5820.6	5998.9
Distribution of TREO %	68.5	28.4	3.0	0.1	100.0	
Distribution of HTREO (Y, Gd, Dy) %	14.5	84.7	0.6	0.2	100.0	

^an.d.: not detected.

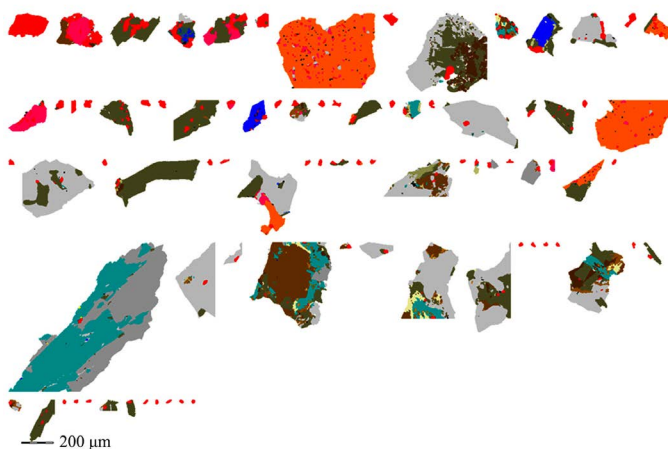
3.2. Target Minerals of REEs

The concentrations of REOs in the ore (**Table 6**) show most (96.9%) of REEs in the ore are carried by monazite (68.5%) and xenotime (28.4%). Thus, monazite and xenotime are the target minerals for beneficiation of REEs. Because xenotime carries 84.7% of heavy REEs (Y, Gd, Dy) in the ore it is more valuable mineral than monazite.

The MLA images of monazite and xenotime particles are shown in **Figure 1**.

Single particles of monazite and xenotime are found in fine grain sizes but many monazite and xenotime particles are associated with other minerals mainly apatite and biotite etc. The grain size (80% passing), the liberation degrees (90% - 95% liberated) and associations of monazite and xenotime measured by MLA are shown in **Table 7**.

Images of monazite particles



Images of xenotime particles



■ Unclassified	■ Quartz	■ Albite
■ K_feldspar	■ Ferrosilite	■ Cummingtonite
■ Gedrite	■ Biotite	■ Muscovite
■ Clinocllore	■ Chamosite	■ Illite
■ Andalusite	■ Cordierite	■ Staurolite
■ Tourmaline	■ Allanite	■ Zircon
■ Calcite	■ Siderite	■ Apatite
■ Monazite	■ Xenotime	■ Wagnerite
■ Magnetite	■ Ilmenite	■ Pyrochlore
■ Masuyite		

Figure 1. MLA images of monazite and xenotime particles.

Table 7. Grain size, liberation and association of monazite and xenotime.

	Grain size, P80 μm	Liberation % (90% - 95% liberated)	Association %				
			Free surface	Apatite	Biotite	Quartz	Monazite Xenotime
Monazite	95	43	48.3	10.5	14.8	9.2	5.0
Xenotime	110	37	50.9	22.5	7.1	4.5	9.3

Table 7 shows that the grain sizes of monazite and xenotime are 95 and 110 μm , respectively, which are appropriate sizes for flotation. The liberation degrees (37% - 43%) are quite low. The free surfaces of two minerals are about 50%. The associated minerals are mainly apatite, biotite and quartz. **Figure 1** shows that monazite and xenotime are associated with these minerals in very fine size.

3.3. Development of Beneficiation Process

The mineralogical analyses above show that monazite and xenotime are the target minerals of REEs in the ore. As phosphate minerals with suitable grain size (around 100 μm) for flotation they could be easily floated with fatty acids or hydroxamic acids as the collectors. For the crushed sample with size of -1.5 mm the liberation degrees (37% - 43%) are quite low. Grinding is necessary for increasing their liberation degrees. Their highly association with apatite, biotite and quartz in very fine size would negatively influence beneficiation efficiency of REEs.

In the basis of preliminary experiments a beneficiation flowsheet was developed (**Figure 2**) which includes grinding, wet low intensity magnetic separation (WLIMS) for removal of magnetite and REEs flotation consisting of one stage of roughing and two stages of cleaning.

Aero 6494, an alkyl hydroxamate developed by CYTEC, was selected by testing as the collector of the REE-bearing minerals. Other conventional phosphate collectors including Na-oleate, EVRR (a fatty acid) mixed with OMC8191 (an emulgator), Aero 845 (a sulfosuccinamate reagent) and S-9849 (an alkyl hydroxamate) were also tested at the grinding size of P80 = 75 μm and neutral pH. Comparing to other collectors Aero 6494 was found to be more selective for the REEs flotation measured by the recovery of REEs (La, Ce, Y) vs. the mass yield as shown in **Figure 3**. S-9849 was also a selective collector, but higher dosage was required comparing to Aero 6494 at the same recovery of REEs.

The effect of grinding size on the flotation performance of REEs is shown in **Figure 4**. In the range of grinding size 75 μm to 180 μm (P80) as the grinding size increasing the REEs (La, Ce, Y) grade of REEs concentrate increases but the REEs (La, Ce, Y) recovery decreases. To get a high recovery of REEs (>90%) at rougher stage the material should be ground to (P80) 75 μm to 120 μm .

3.4. Characterizations of REE Concentrate and Tailings

At the optimized conditions the REEs concentrate was obtained and assayed by

XRF. The mineralogy of the REEs concentrate and the roughing tailings (Tail) was measured by MLA. **Figure 5** and **Figure 6** show that three main REEs, La, Ce and Y, radioactive elements Th and U, and phosphorus (measured by P_2O_5) were significantly enriched in the REEs concentrate and Fe, Si and Mg as impurities were significantly removed.

The mineral grades of apatite, monazite, xenotime, silicate and others in the ore, concentrate and tailings are shown in **Figure 7**. It is seen that phosphates including monazite and xenotime were effectively enriched and silicates as the gangues were removed to the tailings. It is noted that apatite appears in the concentrate at a high grade (65%). Separation of apatite from monazite and xenotime could increase the REE grade of the concentrate. High intensity magnetic separation or acid leaching would be effective approaches for further processing of the concentrate.

The mineral recoveries of apatite, monazite, xenotime, silicate and others in the concentrate, tailings and middling are shown in **Figure 8**. It is seen that although both monazite and xenotime had high recoveries (around 80%) in the concentrate significant amounts of them (14% - 18%) were lost into the middling or the cleaning tailings. Further recovery of monazite and xenotime from the middling by flotation would be challenging because of high concentrations of oxides which could contaminate the flotation of phosphates and high associations of monazite and xenotime with these minerals. Regrinding of the middling may be required. As further investigation in the future a locked cycle test work combined with mineralogical analyses of the cleaning tailings should be performed to study the effect of returning the middling in the flotation process on the flotation performance of REEs.

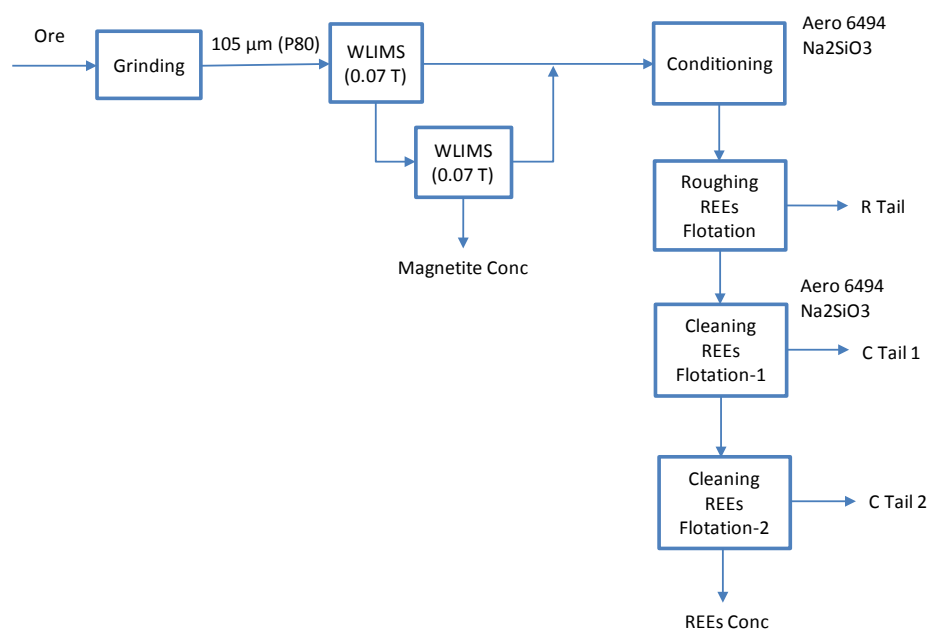


Figure 2. The beneficiation flowsheet for the Olserum ore.

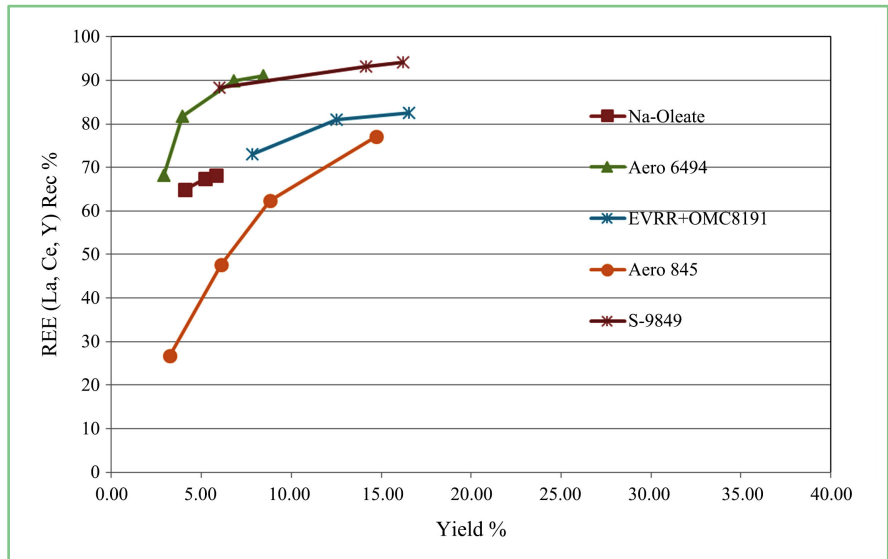


Figure 3. Effect of collector type on the flotation performance of REEs measured by REEs (La, Ce, Y) recovery vs. weight yield of the REEs concentrate (grinding size 75 μm and neutral pH).

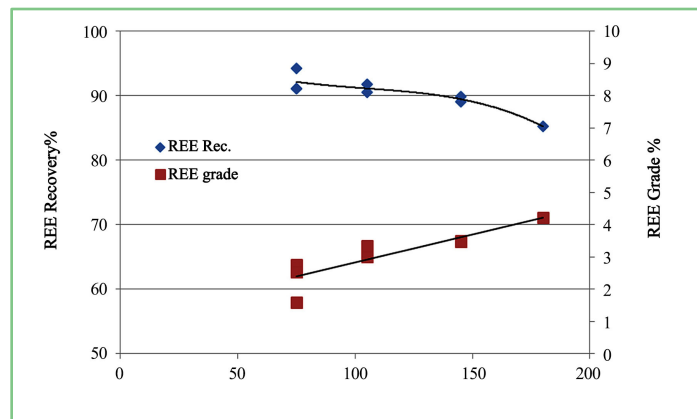


Figure 4. Effect of grinding size on the flotation performance of REEs measured by REEs (La, Ce, Y) grade and recovery of the REEs concentrate (with Aero 6494 as the collector at neutral pH).

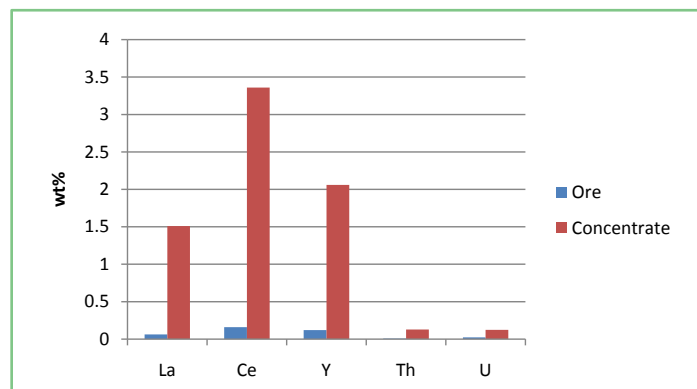


Figure 5. Significance of concentration of REE (La, Ce, Y) and radioactive elements Th and U.

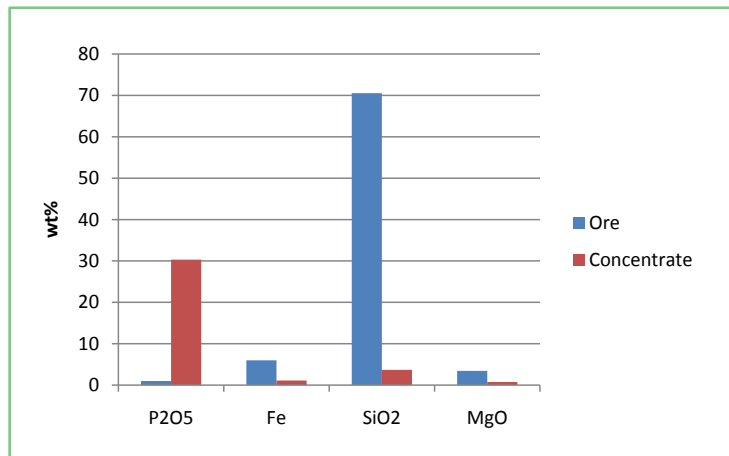


Figure 6. Significance of concentration of P and removal of Si, Fe and Mg.

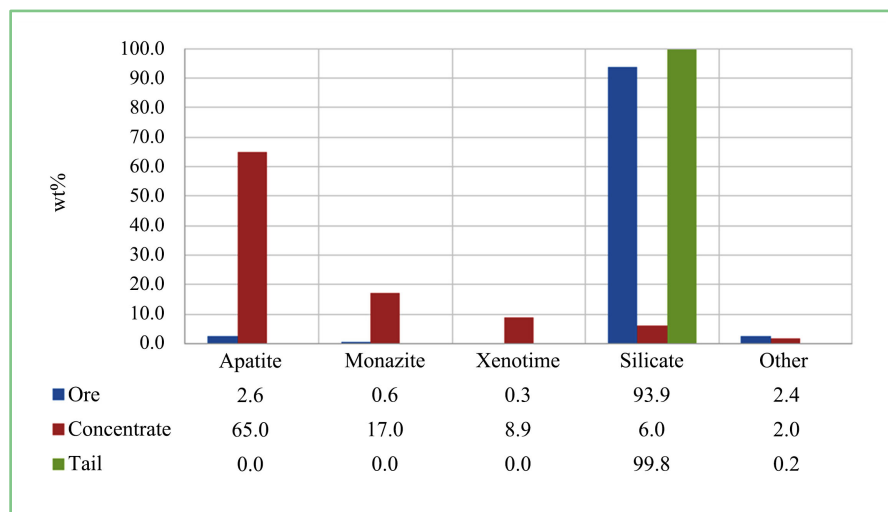


Figure 7. Mineral grades of apatite, monazite, xenotime, silicate and others in the ore, concentrate and tailing.

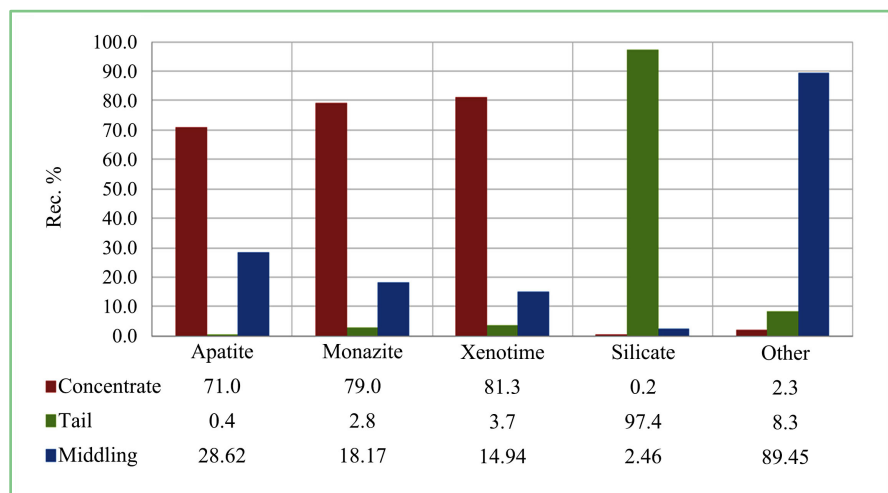


Figure 8. Mineral recoveries of apatite, monazite, xenotime and others in the concentrate, tailing and middling.

4. Conclusions

Chemical analyses show that the content of REEs in the Olserum composite ore is 0.54% and the rate of heavy REEs is 37%. Four minerals were identified by MLA and EPMA to bear REEs, including two phosphate minerals monazite-(Ce) and xenotime, one silicate mineral allanite, and one niobium oxide mineral pyrochlore. Monazite and xenotime are major REE-bearing minerals and carry 68.5% and 28.4% of the REEs in the ore, respectively.

The developed beneficiation flowsheet for the Olserum composite sample includes grinding, wet low intensity magnetic separation (WLIMS) for removal of magnetite and REEs flotation consisting of one stage of roughing and two stages of cleaning. Aero 6494 is more selective than other tested collectors for the flotation of REEs. REE-phosphates display very good flotation behavior in the grinding size range of (P80) 75 μm to 180 μm . As the grinding size increasing the REEs (La, Ce, Y) grade of REEs concentrate increases but the recovery decreases.

Characterizations of REEs concentrate and tailings show that REEs, radioactive elements Th and U, and phosphorus (measured by P_2O_5) were significantly enriched in the REEs concentrate and Fe, Si and Mg as impurities were significantly removed. Monazite and xenotime were effectively enriched and silicates as the gangues were removed to the tailings. Although both monazite and xenotime had high recoveries (around 80%) in the concentrate significant amounts of them (14% - 18%) were lost into the middling or the cleaning tailings. Further recovery of monazite and xenotime from the middling by flotation would be challenging because of high concentrations of oxides and high associations of monazite and xenotime with these minerals.

Acknowledgements

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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