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Study on Processing of Rare Earth Oxide from Monazite, Mongmit Myitsone Region

Aye Thi Tar^a*, Thant Zin Myo^b, Tin Moe Hlaing^c, Bo Bo Mya Win^d

 ^{a,b,c}Material Science Research Department, Department of Research and Innovation, Ministry of Education, Building No.21,Nay Pyi Taw, The Republic of the Union of Myanmar
 ^dAtomic Energy Department, Department of Research and Innovation, Ministry of Education, Building No.21,Nay Pyi Taw, The Republic of the Union of Myanmar
 ^aEmail: dr.ayethitar@gmail.com, ^bEmail: dr.thantzinmyo@gmail.com
 ^cEmail: tmh001@gmail.com, ^dEmail: bbmw2012@gmail.com

Abstract

The present study investigates to support the production of rare earth oxide, which has been used for next research work of individual rare earth elements purification process from monazite, Mongmit Myitsone Region. The processing of rare earth oxide in this study involves four main parts, digestion of monazite concentrate, selective precipitation of rare earth hydroxide, precipitation of rare earth oxalate and calcination of rare earth oxide. The chemicals used in this study were commercial grade from local market. The products from each processing process were characterized by XRD and XRF. The final product contained (> 95 %) of total rare earth oxide and fulfilled to apply next purification process. This paper review the cheaper and easily way to extract rare earth oxide from Myanmar monazite.

Keywords: Calcination; Digestion; Monazite; Rare earth oxide; Selective precipitation.

1. Introduction

The field of the rare earth is fascination. Important research and development work continues globally to explore and establish ways and means to put the rare earths to use, individually and collectively, in the service of humankind.

^{*} Corresponding author.

Rare earth elements have excellent chemical properties which are indispensable in high-tech fields, such as electronic and opt electric and in the production of high intensity magnets and rare earth-nickel based alloys in high capacity nickel metal hydride batteries used in electric-powered vehicles. The term rare earths refer to a group of fifteen elements including those with atomic numbers 57(La) to 71(Lu), as well as yttrium (39) and scandium (21). These elements have varied applications in products of everyday use and also in advanced scientific research [4]. Rare earths are characterized by high density, high melting point, high conductivity and high thermal conductance. A number of rare earth minerals contain thorium and uranium in variable amounts but thorium and uranium do not constitute essential components in composition of the minerals. They are found in combination in mineral deposits widespread throughout the world. Notably large reserves exist in China, the U.S., and Australia. Rare earths in commercially exploitable quantities are found in mineral such as monazite, bastnaesite, cerites, xenotime, gadolinite, fergusonite, allanite and samarskite [2]. Two of the important rare earths minerals produced are monazite and xenotie. Monazite (Ce, La, Th)PO₄ is a phosphate mineral comprises mainly of the light rare earth elements especially Ce, La, Nd and Pr. The mineral also contains considerable amount of heavy rare earths elements notably yttrium and naturally occurring radioactive elements thorium and uranium [4]. In Myanmar, monazite and xenotime occur associated with placer cassiterite and wolframite deposits. Columbite and tantalite also occur in these deposits, which are found in the Dawei and Myeik areas of southern Myanmar [1]. Rare earth oxide can be extracted from monazite and is discovered locally as heavy sands in Myitsone area, Mongmit Township, Homaline area, Sagaing Division, Thabeikyin Township, Heinze and Kanbauk area, Tanintharyi Division and Singu area, Mandalay Division. The location map of regional monazite was shown in Figure1.



Figure 1: location of regional monazite ores

It is widely perceived that the future of the rare earths will be glorious and full of excitement, be it in science, technology, or in commercial utilization. The present study investigates to support the production of rare earth oxide, which was used for next research work of individual rare earth elements purification process from monazite, Mongmit Myitsone Region. The chemicals used in this study were commercial grade from local market. The products from each processing process were characterized by XRD and XRF. This research work involves digestion of monazite concentrate, removing the uranium, thorium and other impurities by using ammonium hydroxide, selective precipitation of rare earth hydroxide, dissolution with nitric acid, purification by oxalic acid and finally oven drying and calcinations of rare earth oxalate to obtain purified rare earth oxide.

2. Experimental procedure

In this study, monazite from Mongmit Myitsone Region, Myanmar was used as raw material. The monazite was physically concentrated from heavy sand in Metallurgical Research Center (ELA), under Ministry of Education. This monazite sand was grounded in a ball mill to finer than 300 mesh size. The flow diagram of the processing of rare earth oxide from monazite concentrate was shown in Figure 2.



Figure 2: processing of rare earth oxide from monazite, Mongmit Myitsone Region

2.1. Digestion process

Rare earth elements are found in combination with mineral deposits widespread throughout the world. They have very similar physical and chemical properties. Therefore their separation processes are extremely difficult. The recovery of mixed rare earth and removal of thorium from monazite are accomplished by variety of methods, after chemically attacking the mineral with sulfuric acid or sodium hydroxide [1]. Because of the

widespread used and commercial process for monazite treatment with caustic soda, sodium hydroxide was used in this process. First, required amount of commercial sodium hydroxide (NaOH), fine monazite concentrate and distilled water were added into the stainless steel beaker (L 335 mm x W 250 mm x H 125 mm) and heated on sand bed. The mixture was agitated slowly by stainless steel rod. The reaction of monazite and sodium hydroxide was shown by equation (1).

$$2 RE(PO_4) + 6NaOH = RE_2O_3 \cdot 3H_2O + 2Na_3PO_4$$
(1)

The mixture was often agitated by stainless steel rod and heated to 140°C. Digestion time was four hours at this temperature. After the mixture was completely digested, the resultant hot mud slurry was cooled down and washed with hot distilled water. All the original phosphorus was present in solution as trisodium phosphate and rare earths and other associated minerals were present as hydrous metal oxide cake. The hydrous metal oxide cake was washed with hot distilled water until all the trisodium phosphate and free sodium hydroxide had been removed from the cake [5].

2.2. Dissolution process

After filtration and washing, filtrate solution was stored as liquid waste. The residues, hydrous metal oxide cake was dried in the drying oven at 110°C for completely dry. The dried cake was grinded with agate motor to make powder form. Then required amount of hydrous metal oxide powder was added into the beaker and placed on a sand bed. After that, slowly added require amount of hydrochloric acid and heated. To completely dissolve the rare earth element, stirred with glass rod gently. The reaction of hydrochloric acid and hydrous metal oxide cake was shown in equation (2). In this process, other associated mineral and rare earths dissolved in acid solution and un-dissolved impurities were left as residue.

$$RE (OH)_3 + 3HCL = RECL_3 + 3H_2O$$
(2)

2.3. Selective precipitation of rare earth hydroxide

The acid solution from hydrochloric acid dissolution process was diluted with hot distilled water and uranium and other associated mineral hydroxide was precipitated at pH-4.5 [7]. The precipitated solution was settled down to complete precipitation. After the filtration, thorium, uranium and other associated mineral hydroxide were stored as solid waste and the filtrate solution was boiled at the sand bed to precipitate rare earth hydroxide at pH-9. After filtration and washed with hot distilled water, residue rare earth hydroxide were dried in the drying oven. The product rare earth hydroxide has white colour.

2.4. Precipitation of rare earth oxalate

After completely dried the earth hydroxide in the drying oven at 110°C and then digested with nitric acid. This mixture was heated on sand bed to complete digest. Then added require amount of hot distilled water to make the solution concentration 100g RE(OH)₃/L and precipitated with 10% oxalic acid. After filtration, the precipitated rare earth oxalate was washed with hot distilled water and oven dried at 110°C. The product rare

earth oxalate has silver white colour.

2.5. Calcination of rare earth oxalate

After completely dried, rare earth oxalate was calcined at 600°C. Then required rare earth oxide was obtained. It has reddish brown colour. The product rare earth hydroxide, rare earth oxalate and rare oxide were shown in figure 3.



Figure 3: rare earth hydroxide, rare earth oxalate and rare earth oxide

3. Results and discussion

The rare earth industry is one of the most fascination and challenging industries in the world. The industry is a fast moving one that is regularly shaken up by swings in supply and demand and the appearance of new applications. Important research and development work continues globally to explore and establish ways and means to put the rare earth to use, individually and collectively, in the service of humankind. The rare earth oxides are the end products of the ore processing and separation operation. In this research work, the composition of monazite concentrate from Mongimit Myitsone Region was characterized by XRD and XRF. The qualitative analyzed of Myanmar monazite sand was shown in Figure 4 and the percentage of the rare earth in the monazite sand was listed in Table 1.



Figure 4: XRD chart of Myanmar monazite, Mongmit Myitsone Region

| Elements | Concentration (%) |
|--------------------------------|-------------------|
| CeO ₂ | 27.206 |
| La_2O_3 | 10.732 |
| Nd_2O_3 | 8.899 |
| Y_2O_3 | 0.887 |
| Pr_6O_{11} | 1.773 |
| Sm_2O_3 | 1.024 |
| Gd_2O_3 | 0.657 |
| Dy_2O_3 | 0.196 |
| P_2O_5 | 19.338 |
| Fe ₂ O ₃ | 1.246 |
| SiO ₂ | 4.257 |
| TiO ₂ | 3.541 |
| ZrO_2 | 0.556 |
| ThO ₂ | 8.164 |
| UO_2 | 0.389 |
| | |

 Table 1: Composition of monazite, Mongmit Myitsone Region

After selective precipitation, rare earth hydroxide was obtained. Because of the co-precipitation of required rare earth elements with solid waste, at least two times selective precipitation was required. The (80-85) % recovery was tested for two times selected precipitation. Oxalic acid is generally the most useful precipitation reagent for the rare earth element, because the separation from other elements is good and the oxalate precipitate can be readily ignited to the oxide [3]. Father purification process followed by selective precipitation was oxalic precipitation and the constituents of rare earth oxalate were listed in Table 2. After calcinations the rare earth oxalate at 600°C for 1 hour, required rare earth oxide was obtained and the purity was also shown in Table 2. The qualitative analyzed of rare earth oxide was shown in Figure 5. Concentration of CeO₂, La₂O₃ and Nd₂O₃ from various processing process were shown in Figure 6, Figure 7 and Figure 8 respectively.



Figure 5: XRD chart of rare earth oxide

| - | | | | |
|------------------|-------------------|------------|----------|--|
| Elements | Concentration (%) | | | |
| | RE Hydroxide | RE Oxalate | RE Oxide | |
| CeO ₂ | 41.070 | 48.973 | 51.530 | |
| La_2O_3 | 23.470 | 25.119 | 25.173 | |
| Nd_2O_3 | 17.05 | 14.173 | 14.215 | |
| Y_2O_3 | 1.384 | 0.709 | 0.547 | |
| Pr_6O_{11} | 3.670 | 3.005 | 3.204 | |
| Sm_2O_3 | 2.46 | 0.802 | 0.737 | |
| Gd_2O_3 | 1.32 | 0.512 | 0.381 | |
| | | | | |

Table 2: Concentration of rare earth oxide at various processing process



Figure 6: Various concentration of CeO₂ in rare earth oxide processing



Figure 7: Various concentration of La₂O₃ in rare earth oxide processing



Figure 8: Various concentration of Nd₂O₃ in rare earth oxide processing

4. Conclusions

Rare earths in the Myanmar Monazite, which was located at Myitsone Area, Mongmit Township, can be separated and purified by chemical process and calcined to convert rare earth oxide for support individual rare earth elements purification. The monazite concentrate was digested with sodium hydroxide. In digestion process, although various ratios of monazite concentrate to sodium hydroxide ratio of (1:1.5), (1:1.75), (1:2), (1:2.25), (1:2.5) and (1: 2.75) were studied. Among them, the ratio of 1:2 is suitable and, digestion time of four hours and temperature 140°C were fulfilled the require condition. In hydrochloric acid dissolution, 1 kg hydrous metal oxide cake per 1 L (1kg/1L) concentration was suitable. For selective precipitation, uranium, thorium and other associated minerals hydroxide were separated at pH-4.5 and rare earth hydroxide was precipitated at pH-9. Nitric acid dissolution was also suitable for ratio (1:1) of rare hydroxide to nitric acid. After precipitation with 10 % oxalic acid, rare earth oxalate was obtained and calcinations at 600°C for one hour to produce final product rare earth oxide. The final product rare earth oxide was characterized by XRD, XRF and it contained CeO₂ (40-52)%, La₂O₃ (20-25)%, Nd₂O₃ (12-20)%, Pr₆O₁₁ (3-4)%, Sm₂O₃ (2-3)%, Y₂O₃(1-2)% and average (>95%) of total rare earth oxide. It was successfully completed by using the chemicals which were easily supplied from local market and the products fulfilled to apply next purification process.

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