Properties of Rare-Earth Element in Magnetic Material and Its Processing

Rare-earth metal is one of the critical elements because of its small amount with a lot of demand for this metal in a variety of the latest technologies, which are currently developed fast and intensively. The use of rare-earth metals can contribute to the development of innovations in the production of new materials in various fields, because these metals have strong, hard and heat-resistant properties. Rare-earth metals are found in complex compounds that makes it difficult to separate from ore. Obtaining of the rare-earth metals is realized by recycling the product from a secondary source of magnets containing rare-earth metals, such as NdFeB and SmCo magnets. Hydrometallurgical and pyrometallurgical processes can carry out the process of recovering of the rare-earth metals from secondary materials. There is a new research using bacteria as a rare-earth metal extractor to minimize environmental impact. Oxalic acid and other organic acids have potential in the recovery of rare-earth metals. As alternative, the rare-earth-free materials as candidates for permanent magnets are also mentioned.

Keywords: rare-earth elements, magnetic material, processing of magnetic materials.

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1. Introduction

The rare-earth metal is a lanthanon metal, which has atomic numbers from 57 to 71 (15 elements). This metal consists of lanthanum-to-lutetium elements, and there are additional two elements, namely, yttrium and scandium. The addition of these two elements makes the rare-earth metals group based on their acidity [1]. This metal is called a rare-earth metal because it took a very long time to find this element, which was almost 160 years around 1788 to 1941. Rare-earth metals can be grouped based on atomic mass, atomic weight, ionic radius, electron arrangement, and the process by which they occur.

Each rare-earth metal group element has the same acidity properties so that these elements can be found in the same minerals. Rare-earth metals have several properties, namely that this element can react with water and oxygen, is a stable compound when in the form of an oxide, has a relatively high melting point and is a good thermal conductor [2]. Rare-earth metals can react exothermically with the element’s selenium, sulphur and phosphorus. Rare-earth metals are soluble in dilute acids and hydrogen, sparingly soluble in concentrated sulphuric acid solutions. Rare-earth metals react with organic acids more slowly than mineral acids with higher concentrations [1].

Rare-earth metals are not found in free form in nature, but as complex compounds of phosphates and carbonates or bonded to mineral compounds. The minerals lime, zircon, calsterite, and monazite are minerals that contain many rare-earth metals. In the mineral monazite, it was found about 50% to 67% rare-earth metals [3]. The use of rare-earth metals cannot be used directly, but through the process of separating elements from their complex compounds. This separation is carried out based on the magnetic and electrical properties of the minerals. The use of rare-earth metals can develop innovations in the production of new materials in various fields because these metals have strong, hard and heat resistant properties. Rare-earth metals are also widely used in everyday life such as in steel plates, automatic gas lighters, security lights, jewellery, paint and glue [4]. The unique physical and chemical properties of rare-earth metals make them high quality materials in a variety of advanced equipment. The increasing demand for this element and the difficult extraction process make this element very rare in the market. Reduced supplies of rare-earth metals create environmental problems for the primary rare-earth metal industry, which makes research and development in the processing of secondary resources derived from mining waste and electronic equipment solid waste. Extraction of rare-earth metals from waste is one of the solutions to environmental problems and guarantees sustainable production of rare-earth metals in the future [5].
Many developed countries import rare-earth metals from China. About 95% of rare-earth metals are exported from China, thus increasing the export price from China due to the large quota of rare-earth metals supplied by China [6]. Although research to extract rare-earth elements from secondary sources has been realized and there are developments with other technologies, most are only suitable for the extraction of some secondary sources. In metallurgical processing, leaching becomes a step that can be used for rare-earth metals from residues, minerals, or waste containing rare-earth metals.

2. Magnetic Materials

2.1. Neodymium–Iron–Boron Magnet

NdFeB is a type of permanent magnet that has the best quality compared to other types of permanent magnets known since 1984. NdFeB magnets have superior energy (with a maximum energy of 512 kJ), thus, making this type of magnet very efficient and suitable in various applications of lightweight portable products [7]. Therefore, NdFeB magnets are widely used in electronic goods such as wind turbines, hybrid and electric vehicles, computer hard disks (HDD), and many more. These permanent magnets have different life cycles based on their use, such as in consumer electronics, their life cycle is 2–3 years, while in wind turbines, these magnets have a longer life cycle of 20–30 years. In consumer electronics equipment, NdFeB magnets used are less than 1 gram and reach 1000–2000 grams in wind turbine generators. In these magnets, there are about 31–32% rare-earth metals, of which the elements Nd and Pr are most abundant, with small but expensive amounts of Dy, Tb, and Gd. Recycled rare-earth metals can be used in end-of-life (EOL) magnet types, which play an important role in fulfilling rare-earth supplies in the future. However, the recovery of small consumer electronics magnets is experiencing many social problems and great challenges. At present, there is no efficient technology, which can be used to recycle rare-earth metals. It is hoped that by 2030 the recycling of rare-earth metals from EOL permanent magnets will have an important role in the supply of rare-earth metals [8].

More than 20 types of known applications contain rare-earth metals [9]. Figure shows the magnetic applications of rare-earth metals [10]. Applications for rare-earth magnets that play a major role are hard disks and CV/DVDs. As you can see, motors and generators are one of the applications with the highest use of rare-earth metals in recent years. Even though these data are old and different from other sources in recent years, some similarities can be observed. Grouping based on different applications will cause inconsistent data obtained.
2.2. Samarium–Cobalt Magnet

SmCo alloys are magnets that come from a combination of samarium and cobalt elements. Compared to NdFeB magnets, the processing of these magnets is easier because the SmCo powder does not react. Therefore, the development of SmCo magnet pre-processing is focused on the grinding and crushing steps [11]. According to Ref. [12], the thing that must be done is a treatment to avoid binding of magnetic material during the washing process is the demagnetization process. In addition, heating at high temperatures can change Sm into oxide compounds. Demagnetization is preferably carried out after the grinding process at 850°C for 6 hours under atmospheric pressure, so that Sm oxide compounds can be produced from thermogravimetric analysis and x-ray diffraction analysis, which provide information that there is no SmCo alloy [12]. The milling process can affect the efficiency of the washing process. Better Sm recovery 75–95% with reduced particle size from 125 to 74 m. However, a decrease in particle size that is more than this seems to be unfavourable and does not affect the Sm leaching process [13]. The literature on the pre-processing of SmCo magnets is very limited, so demagnetization at temperature is recommended 850°C and particle sizes below 150 m to provide higher recovery rates.

Hydrometallurgical leaching processes using acid solutions after demagnetization and grinding of SmCo magnets have been studied recently. In this process, almost 100% recovery of Sm was achieved under optimal conditions at 4 M concentration with HCl solution, 500 rpm, 10% S/L at room temperature 95°C for 120 minutes. However, this method did not show the selectivity of Sm extraction even though the co-extraction yield was high at around 90% [11]. Authors of Ref. [14]...
studied the leaching parameters with the results obtained similar to those of previous research, namely with a concentration of 4 M and 17% S/L at 60 °C for 75 minutes. However, Sm recovery rate was relatively lower, namely 24.1% and 99.9% for Co. Other authors [13] carried out leaching to extract Sm and Co from cobalt-based threshing magnets at 80 °C for 1 hour with 10% S/L. Resulting in high Sm and Co recovery rates with 88.5% and 95.4%, respectively. However, this method is not selective in extracting Sm because impurities such as Fe are found in the solution. In another study, it was found that hydrochloric acid and sulphuric acid were more efficient in extracting Sm from SmCo magnets with recovery rates of 95% and 79%. High recovery efficiency is achieved at an S/L ratio of 50 g/L at 80 °C for 120 minutes [15]. The hydrometallurgical process is a non-acid dissolving process using copper (II) salts, especially Cu to dissolve rare-earth metals from magnetic materials. Ionic copper could oxidize and dissolve rare-earth metals with copper metal or copper oxide as a by-product. A hydrometallurgical method to recover Sm from SmCo through a demagnetization process has been proposed in Ref. [16]. Research on the extraction of Sm from SmCo is very limited and focuses on the use of inorganic acids or HCl solutions in the leaching process.

3. Hydrometallurgical Processes

The hydrometallurgical process in rare-earth metal extraction consists of several steps, namely magnetic washing, separation of rare-earth metal elements (Nd, Pr, Dy) using solvent extraction through a leaching process, ion exchange through ionic liquid extraction techniques. Precipitation of rare-earth metal elements from non-rare-earth element specimens is carried out which is dissolved together and converted to fluoride or rare-earth metal oxides. Recovery of rare-earth metals is a challenge in hydrometallurgical processes especially in the selective use of wastewater, which can dissolve, increase concentration, and separate rare-earth elements from their alloys and complete recovery of all rare-earth metals at one time. Therefore, it is necessary to use separation technology using new extraction solvents, ionic liquids, and other processes.

3.1. Acid Leaching

The first step in the hydrometallurgical process is dissolving the rare-earth metals through a magnetic leaching process. The method of decomposing waste magnets can be done in three ways, namely: (1) magnetization/demagnetization of magnets (with or without burning), (2) combustion by washing rare-earth metals and (3) converting/changing magnets containing rare-earth metals into magnets by new solid phase.
The conversion of rare-earth metals in magnets into precipitated rare-earth metal compounds can be carried out based on the solubility of rare-earth metals at different temperatures under hydrothermal conditions. A magnetic washing method at room temperature has been developed in Ref. [17]. Nd can be precipitated as a sulphate salt, which can turn into NdFe₃. However, most of the Fe is also dissolved; thus, it must be removed properly. Authors of Ref. [18] successfully applied filtration in EOL processing of NdFeB magnetic complexes from HDD. As demonstrated in Ref. [19], a washing process with HCl selectively dissolves NdFeB magnets from an electric motor. Magnetic material demagnetization was carried out using 4 M HCl solution for 24 hours at room temperature, while steel and copper did not react at all. Then, oxalic acid is made as a leach solution to precipitate rare-earth metal retreat (Nd, Pr, and Dy) [18].

3.2. Solvent Extraction

The separation of solvent extracting ions from solution is carried out based on the formation of complex compounds between dissolved ligand molecules in the organic and ionic phases present in the aqueous phase to transfer the complex compounds to the immiscible phase. The distribution of metals is affected by the chemical equilibrium between the metal forms in the two phases and the free energy changes occur when the metal-containing specimen is transferred from one step to another one. This process depends on chemical properties, separation, ligands, solvents, pH of the aqueous phase, temperature, and others.

The separation of the lanthanides from actinium is very important in the nuclear industry. One of the difficulties in separating rare-earth metals is due to the chemical nature of these elements [20]. These elements have almost similar properties; the similarity of properties can affect the formation of complex compounds in the liquid phase, which results in separation efficiency using solvent extraction. The difference in the electron nuclear charge between the ions must be used in the joint separation of the ions [21, 22]. In general, the use of pure rare-earth metals from pure ores includes several stages of mixing precipitation to give a yield of sufficient purity.

3.3. Ionic Liquids

The use of organic substances in solvent extraction is a novelty in hydrometallurgical processes. Ionic liquids are liquids that are composed of ions. Ionic liquids are considered safer in replacing molecular organic solutions in the extraction process of separating rare-earth metals because they are non-volatile and non-flammable [23–27]. Very high concentrations of extracts in ionic liquids can act as base extracts to
replenish metals in the liquid phase and enhance the process. There are several challenges in using ionic liquids for the solvent extraction process, including: (1) ionic liquids have a high viscosity thereby inhibiting mass transport, (2) extracting using ionic liquids causes ion exchange, resulting in the loss of the aqueous phase composition in ionic liquids, (3) the hydrolysis instability of some anions, (4) ionic liquid recycling, (5) toxicity problems, (6) has a high price. One example of the use of ionic liquids in the separation of rare-earth metals is using a combination of liquid ions and their extracts [31]. N,N-dioctyldiglycolamic acid (DODGAA) is a modification of tetraoctyl diglycolamide (TODGA) extract, which has a strong affinity. In this system, rare-earth metals are extracted as 1:3 complexes. Stripping is a problem that complicates the use of ionic liquids such as TODGA [28–30]. Kikuchi et al., [31] conducted an experiment to extract Pr, Nd, and Dy ions with tributylphosphate. One example of the use of ionic liquids in the separation of rare-earth metals is using a combination of liquid ions and their extracts [31]. DODGAA is a modification of TODGA extract, which has a strong affinity. In this system, rare-earth metals are extracted as 1:3 complexes. Stripping is a problem that complicates the use of ionic liquids such as TODGA [28–30]. Kikuchi et al., [31] conducted an experiment to extract Pr, Nd, and Dy ions with tributylphosphate. One example of the use of ionic liquids in the separation of rare-earth metals is using a combination of liquid ions and their extracts [31]. DODGAA is a modification of TODGA extract, which has a strong affinity. In this system, rare-earth metals are extracted as 1:3 complexes. Stripping is a problem that complicates the use of ionic liquids such as TODGA [28–30]. Authors of Ref. [31] conducted an experiment to extract Pr, Nd, and Dy ions with tributylphosphate.

4. Pyrometallurgical Processes

The main objective of the pyrometallurgical method is to use high temperatures to convert rare-earth metal magnets into rare-earth metal elements separated by different phases. The rare-earth metals are separated into other phases, which are so concentrated that they can be produced by electrolysis of molten salts. The pyrometallurgical method is divided into several steps, namely heating, extracting molten metal, extracting molten salts, and electrochemistry. Descriptions of the recovery of rare-earth metals from magnets at high temperatures can be found in several recent studies [32–35]. Authors of Ref. [32] emphasized that processing at high temperatures is an alternative way to avoid consuming a lot of water and producing hazardous waste in the recovery process.
4.1. Roasting

The purpose of baking is to change the shape of the magnet at high temperatures. After the roasting process, it is expected that a more efficient separation process be achieved in the metallurgical industry. For example, in sulphate roasting, during the sulphide roasting process, the metal will be sulphated to form water-soluble sulphates. This process consists of two steps [36], in the first step, the magnetic material is converted to sulphate by decomposition into a solid state at room temperature, and the resulting mixture is heated so that the rare-earth metal sulphates remain thermally stable. Roasting with sulphates, then washing with water removes 95% of the rare-earth metals during leaching of the iron-free solution. Iron produces a by-product as hematite, which can be used as a pigment. Impurities such as Co and Ni are also present in the residue enabling the production of leaching with a purity of 98%. In this way, the recovery of rare-earth metals can be developed and can minimize emissions. The purpose of oxidizing roasting is to convert rare-earth metal magnets into oxides so that the subsequent leaching process is easier, faster and more selective [37–41].

Ames laboratory of liquid metal extraction (U.S.A.) developed the recovery of rare-earth metals through a process of selective liquid metal extraction from NdFeB magnets and other magnets containing rare-earth metals using Mg as an extractant [42], which led other researchers to study this process [43–45]. Nd, Pr and Dy can be selected at 1000°C, leaving Fe–B and other unreacted transition metals in the residue. Moreover, selective extraction of Nd was successful using silver [46] and copper [47]. By using Ag as an extraction solution, Nd–Ag containing 45–50% Nd is obtained at a temperature of 1000–1300, and Nd can be separated by oxidizing Nd–Ag, which is liquid in air. As argued in ref. [48], a magnetically molten copper alloy (around 1–2 min), and a Cu–Nd-rich intermetallic phase is formed on cooling of 350 °C/min.

4.2. Direct Electrochemical Filtration

Electrochemical processes can be used on magnetic metals with the principle of purification using molten salt [35]. In electrolytic cells, a piece of impure magnet is used as the anode and the rare-earth metals can be selectively dissolved in liquid chloride or fluoride, while for the cathode it is preferable to use rare-earth metals. The management of waste magnets at high temperatures has been evaluated in Ref. [34] by describing the advantages of dry metallurgical processes over wet ones. The challenges in recovering rare-earth metals from the used magnets are as follow [35]: (1) differences in the waste mixture or waste composition, (2) the effect of pollutants on the recycling process, (3) optimization in
the separation of rare-earth metals (Nd, Pr, and Dy), and (4) the economic feasibility of the recycling process and the life cycle of the magnet.

5. Green Processes

In the recovery of rare-earth metals, there are many environmental pollution problems, which occur in the form of toxic emissions, excessive amounts of solid and liquid waste and the presence of radioactive materials. For this reason, efforts are being made to find new methods for sustainable recovery of rare-earth metals, but with minimal environmental hazards. Some of the developments in reducing harmful emissions are libenesit treatment, biological cleaning, and the use of microwaves in the washing process.

5.1. Use of Bastnaesites

Environmental pollution due to the roasting process is getting more and more attention [49]. Therefore, a process was developed to prevent fluorine emission. The first method was developed to leaching only rare-earth metal carbonates by allowing fluoride in the residue. It is obtained at a temperature of 400 °C for 3 hours and then washed with HCl solution. The resulting efficiency is 0.07% for fluoride and 94.6% for carbonate [49]. There has not been an effective way of dissolving fluorinated compounds, but changes in oxidation of Ce³⁺ to Ce⁴⁺ can prevent Ce from joining with the rare-earth metals. The addition of thiourea [50] can be used to overcome this problem; thiourea can prevent Ce oxide after roasting and allows washing of Ce with HCl so that recovery of rare-earth metals is not bound to fluoride. This method is not optimal because not all of the free site rare-earth metals are extracted; so, it is less efficient. Another more efficient process with mechanical crushing of liberation uses NaOH powder [51].

5.2. Bioleaching

At present, materials are being explored in the field of bioreactors to extract rare-earth metals from resources with low energy levels. The resources in question are old ores and ion-absorbing clays. Due to complete leaching, the use of strong acids and bases generates large amounts of waste in an inefficient, cheap, and clean manner. In Egypt, the bacterium Acid thiobacillus ferroxidase has been used for bioleaching low-grade gibbsite ore to recover rare-earth metals (0.49%) and uranium (0.05%), which is generally used in copper leaching [53]. Other types of bacteria tested were Aspergillus ficuum and Pseudomonas aeruginosa, whereby the use of these bacteria as a rare-earth metal wash resulted in an efficiency of about 75% [54].
5.3. Microwave

Microwave is often used to increase mineral leaching efficiency [55]. The principle of microwave-assisted leaching is based on the fact that metal carrier minerals are less transparent to microwaves than CaO compounds [55]. This refers to local heating which can change the leaching kinetics. In general, the leaching kinetics increases with increasing temperature allowing faster leaching of metal minerals and less absorption of unwanted specimens. Another impact that occurs from fast heating is that it can damage the surface of metal minerals due to thermal stress thereby increasing the surface area [56]. For example, removal of the reaction product from the surface via metal-containing (Cu) or (Au) convection currents would be beneficial in rare-earth metal leaching. It can be considered that with current applications, microwaves can be applied in the leaching of rare-earth metals, especially sources with low energy levels. Interest in this method is based on the success of microwave (Cu) and gold (Au) leaching of chalcopyrite [56].

6. Rare-Earth-Free Candidates for Permanent Magnets

As currently known [57, 58], permanent magnets are one of the strategic metal-based products in the world industry due to a wide range of their applications: from micro-electromechanical and nano-electromechanical systems to high-power electricity generators using many tons of magnetic materials containing critical rare-earth metals. Such non-renewable elements are the main factor that constrains or increases the cost of fabrication of magnetic products. Therefore, the task of obtaining new permanent magnets, which have comparable characteristics to existing magnets, but free from critical elements (such as rare-earth metals), is extremely important for industrial and national securities.

Recent years a great progress in the world has been made toward studying and feasible improving the microstructure and physical properties of non-rare-earth materials for permanent magnets [57, 58]. Researchers investigated several new materials-candidates. Some of them have showed realistic potential for replacing rare-earth permanent magnets for some applications [57, 58]. Properties of these materials are described in dozens and even hundreds of published research articles and several reviews. In the recent review [58], authors addressed about such systems as Mn-based Mn–Al and Mn–Bi alloys with high magnetocrystalline anisotropy, spinodally decomposing Fe-based Al-, Ni-, Co-containing (Alnico) alloys, high-coercivity tetrataenite $L1_0$-phase in Fe–Ni [59–65] and Fe–Co [66, 67], Co-rich HfCo$_5$ and Zr$_2$Co$_{11}$ intermetallic compounds, Co$_5$C and Co$_3$C carbides, and still mysterious iron nitride $\alpha''$-Fe$_{16}$N$_2$ phase. The latter system attracts a considerable interest [68–70] due to its exceptionally high saturation magnetization, low cost of
Fe, and because its elements are the most earth abundant among all magnetic materials.

7. Summary

Rare-earth metals are one of the critical elements because of their small amount with a lot of demand for such metals in a variety of the latest technology, which currently develops rapidly. Rare-earth metals are found in complex compounds making it difficult to separate from ore. To obtain the rare-earth metals, it is realized through recycling the product from a secondary source of magnets containing rare-earth metals such as NdFeB magnets and SmCo magnets. Compared to other types of permanent magnets, NdFeB magnets are rated as the best magnets containing rare-earth metals. Hydrometallurgical and pyrometallurgical processes can carry out the process of recovering rare-earth metals from the secondary materials. The hydrometallurgical process is the most widely used process, because it has several advantages over pyrometallurgical processes. The use of these two methods still causes a high environmental impact, so currently processing of rare-earth metals is being developed using organic matter in the leaching process, such as the use of glycolic acid, ascorbic acid, maleic acid, citric acid, acetic acid, oxalic acid and other organic acids which have potential in the recovery of rare-earth metals. In addition, there is new research using bacteria as a rare-earth metal extractor to minimize environmental impact. Oxalic acid and other organic acids have potential in the recovery of rare-earth metals. There is new research using bacteria as a rare-earth metal extractor to minimize environmental impact.

The new tailored metallic phases do not containing critical elements (non-renewable on the Earth) may act as an alternative to the permendur and rare-earth permanent magnets. Researchers investigated several new materials-candidates. Some of them have showed realistic potential for replacing rare-earth permanent magnets for some applications.

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ВЛАСТИВОСТІ РІДКІСНОЗЕМЕЛЬНОГО ЕЛЕМЕНТУ В МАГНЕТНОМУ МАТЕРІАЛІ ТА ЙОГО ОБРОБЛЕННЯ

Рідкісноземельний метал є одним із найважливіших елементів через його невелику кількість з великим попитом на цей метал у різноманітних новітніх технологіях, які наразі швидко й інтенсивно розвиваються. Використання рідкісноземельних металів може сприяти розвитку інновацій у виробництві нових матеріалів у різних галузях, оскільки ці метали мають міцнісні, твердотільні
та термостійкі властивості. Рідкісноземельні метали містяться у складних сполуках, що утруднює виділення їх з руди. Одержання рідкісноземельних металів реалізується шляхом переробки продукту з вторинного джерела магнетів, що містять рідкісноземельні метали, наприклад магнети NdFeB і SmCo. Процес відновлення рідкісноземельних металів із вторинних матеріалів може здійснюватися за допомогою гідрометалургійних і пірометалургійних процесів. Є нові дослідження з використанням бактерій як екстрактора рідкісноземельних металів для мінімізації впливу на навколишнє середовище. Щавлева кислота й інші органічні кислоти мають потенціал для відновлення рідкісноземельних металів. Як альтернативні згадуються також матеріали-кандидати для постійних магнітів без вмісту рідкісноземельних елементів.

Ключові слова: рідкісноземельні елементи, магнітний матеріал, оброблення магнітних матеріалів.