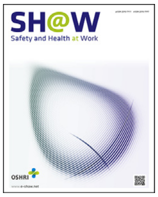
Safety and Health at Work 10 (2019) 409–419

Contents lists available at ScienceDirect

Safety and Health at Work

journal homepage: www.e-shaw.net



Review Article

# Worker Safety in the Rare Earth Elements Recycling Process From the Review of Toxicity and Issues

Seo-Ho Shin, Hyun-Ock Kim, Kyung-Taek Rim*

*Chemicals Research Bureau, Occupational Safety and Health Research Institute, KOSHA, Daejeon, Republic of Korea*

ARTICLE INFO

*Article history:*
Received 9 April 2019
Received in revised form
16 August 2019
Accepted 26 August 2019
Available online 31 August 2019

*Keywords:*
Hazard/risk
Occupational safety and health
Rare earths
Recycling
Toxicity

ABSTRACT

Although the rare earth elements (REEs) recycling industry is expected to increase worldwide in high-tech industry, regulations for worker safety have yet to be established. This study was conducted to understand the potential hazard/risk of REE recycling and to support the establishment of regulations or standards. We review the extensive literature on the toxicology, occupational safety, and health issues, and epidemiological surveys related to the REEs, and propose suitable management measures. REE recycling has four key steps such as collection, dismantling, separation, and processing. In these processes, hazardous substances, such as REEs-containing dust, metals, and chemicals, were used or occurred, including the risk of ignition and explosion, and the workers can be easily exposed to them. In addition, skin irritation and toxicities for respiratory, nervous, and cardiovascular systems with the liver toxicity were reported; however, more supplementary data are needed, owing to incompleteness. Therefore, monitoring systems concerning health, environmental impacts, and safety need to be established, based on additional research studies. It is also necessary to develop innovative and environment-friendly recycling technologies, analytical methods, and biomarkers with government support. Through these efforts, the occupational safety and health status will be improved, along with the establishment of advanced REE recycling industry.

## 1. Introduction

The term “rare earth elements” (REEs) means that they are rarely found in nature. Strictly speaking, this is incorrect because the term “rare earth” derives from the minerals from which they were first isolated, which were uncommon oxides found in gadolinite extracted from a mine in the village of Ytterby, Sweden. Actually, excluding promethium, rare earth elements are diffusely found in the earth’s crust. Because the REEs are generally chemically stable and have excellent thermal conductivity, high ductility, and corrosion resistance, they are widely used as a core material in electric and hybrid vehicles, wind turbines, solar power, optical fibers, phosphors, displays, catalysts, medical industries, and structural alloy steel, and their usage and price are increasing. Recently, REEs have been recognized as an essential resource for the transition to high-tech industries and are attracting worldwide attention as the vitamins of modern technology [1].

Although the ore reserves of REEs are not too small, like monazite and bastnasite, their mines are limited to specific counties. In addition, they are mainly produced in China, owing to bioaccumulation, human toxicity, and environmental pollution caused by dust, metal, radioactive material, organics, and wastewater generated during mining, purification, and separation [2,3]. Other productive countries, such as Australia, the United States, and Russia, have reduced their production scale before, owing to technical or environmental reasons. Despite having a few reserves, there is no production of REEs in Korea, considering their economic efficiency [4]. China holds about 55 million tons of REEs that are the largest reserves in the world (48.4%) and produced more than 80% of the world’s production of rare earth oxides (REOs) in 2016 (about 155,000 tons) [5]. For that reason, REEs were regarded as critical raw materials in the 2010 annual report of the European Union [6]. The US Department of Energy defined dysprosium (Dy), neodymium (Nd), terbium (Tb), europium (Eu), and Y as critical in the short- and mid-term, considering the supply risk and their importance to

---

* Corresponding author. Occupational Safety and Health Research Institute, KOSHA, Daejeon, Republic of Korea.
*E-mail address:* rim3249@gmail.com (K.-T. Rim).

https://doi.org/10.1016/j.shaw.2019.08.005

clean energy technologies [7]. In Korea, REEs were classified as some of 35 rare metals, and their depletion is expected in the future with the development of the new growth engine of Korean industry in the 21st century.

Because of the scarcity of REE resources and technical/environmental barriers, China monopolizes the global REE market and uses them diplomatically as an offensive tool; therefore, many client countries have had difficulty in the domestic supplies of REEs. In particular, Japan was forced to have interest in the development of permanent magnet recycling technology as a self-rescue measure, in response to an import prohibition of REEs that resulted from a territorial dispute with China in 2010. In addition, China caused a price jump and imbalanced supply between heavy rare earths and light rare earths, by reducing the export quota to cope with rapidly increasing domestic demands [8]. Consequently, Europe and Japan began to activate the recycling industry for the stable supply of REEs. In the case of Japan, the recycling rate of REEs was only 0% in 2011, but it is now consolidating its position as a powerful recycling country through political support and continuous research/development [3]. Korea also became interested in REE recycling from e-wastes, but scraps containing Nd and Dy, which are the main components of permanent magnets, were still disposed of or exported, despite the more than 50% collection rate [9].

Industrial health issues are becoming problematic as the REE industry develops. It is well known that pulmonary fibrosis and pneumoconiosis were observed in workers chronically exposed to REEs. The *in vivo/in vitro* toxicities of REEs supporting these issues have also been reported [10–13]. Various harmful substances have been emitted from REE mining to processing, and REE recycling can also cause health and environmental problems, owing to the similarity of the process. Dillow (2011) reported that only 10%–15% of personal electronic devices globally were properly recycled [14]. As described previously, while REE problems and issues for living organisms and local environments have been on the rise, their regulations and managements are still insufficient [3]. Only Y and cerium (Ce) in the workplace are managed as 1 and 5 $mg/m^3$, respectively, but more compatible regulation is necessary because handling forms of REEs may vary from raw materials to finished products, and the substances generated may also be different for each process [12].

As the REE industry has recently developed, the importance of international standardization for terms, recycling, environmentally friendly technology, etc. is highlighted, and the REEs technical committee (ISO/TC 298) was newly established in 2015. This technical committee that consists of 34 countries, including Korea, has five working groups (WGs) up to now, aimed at establishing universal standards for various industrial activities and standards for environment-friendly technology. Korea deals with working group 2 (WG2; recycling), and the expert committee led by the Korean Agency for Technology and Standards (KATS) and the Korea Institute for Rare Metals (KIRAM) was established, to keep up with the global trend. Therefore, according to the international prospect for REE recycling and organization of the ISO/TC 298, this research tried to suggest measures for worker safety in the upcoming REE recycling industry.

## 2. Materials and methods

To understand the effects on human and environment during the REE recycling and to prepare measures for the related worker safety, this research organized and reviewed various publications and articles about REE recycling and technologies, occupational safety and health issues, epidemiological survey, *in vivo* and *in vitro* toxicities, human toxicity, and environmental pollution. Online media, mainly Google (https://www.google.co.kr), Google Scholar (http://scholar.google.com), Scopus (https://www.scopus.com), Science Direct (https://www.sciencedirect.com), NDSL (http://www.ndsl.kr/index.do), Pubmed (http://www.ncbi.nlm.nih.gov/pubmed), and KOSHANET (http://www.kosha.or.kr/bridge?menuId = 69), were utilized to collect the data. Searching was performed by using the key words of “rare earths,” “recycling industry,” “recover,” “collection,” “urban mining,” “rare earths containing wastes,” “permanent magnet,” “nickel metal-hydride batteries (Ni-HM),” “lamp phosphors,” “hydrometallurgical methods,” “pyrometallurgical methods,” “hazardous substance,” “environmental pollution,” “toxicology,” “exposure limit,” and “occupational safety and health issues.”

To accomplish the purpose of this research, only necessary data were selected from the searched data, and it was then summarized into the following categories: “REEs recycling and usage status,” “Hazard/risk of REEs recycling,” “Toxicity and occupational safety and health issues of REEs,” and “Requirements for worker safety in REEs recycling.” Based on these summaries, measures for worker safety and the future direction of the REE recycling industry were discussed.

## 3. Results

### 3.1. Recycling of REEs and usage status

Recycling processes of REEs can be mainly divided into four key steps such as collection, dismantling, separation (preprocessing), and processing as other metals. The collection as the first step can be performed by various methods, such as transporting expired products to a designated place or a collection area near the manufacturing factory. This step is easier to perform because policy support and public infrastructure are well established. In the case of Japan, which is an advanced recycling country, the collection routes for REE-containing wastes are systematically established with the development of the urban mining project. Dismantling and separation are mostly performed by manual and mechanical processes, and their methods are applied depending on the type of waste products. Hitachi developed the process using a rotating drum for disassembling hard disk drives, and the National Institute for Materials Science (NIMS) developed a small-scale electronic crushing device and three-dimensional ball mill, resulting in sharply shortened work time [3,15]. Processing generally consists of pyrometallurgy, hydrometallurgy, electrochemical process, and materialization technology, and recently, these methods have tended to be combined. However, REE recycling is still not practical because current recycling technologies are not yet able to meet the three factors of “economics,” “technology,” “society,” which are essential for efficient recycling. For example, recovery of rare earth metals (REMs) from molten scraps shows low efficiency, and recovery as REOs also has a low price on the market. Currently, the application fields of REEs are limited to permanent magnets, polishing agents, batteries, and lamps, and commercialized cases are very few, compared to many research achievements [16–18]. Nonetheless, many countries have continuously tried to develop REE recycling technologies, because of the inexhaustible possibilities and future-oriented aspects of REEs resources.

In the fields of magnets, recycling methods are mainly divided into three methods: extracting/recovering REEs by the smelting process, recycling as a magnetic alloy material, and the reuse of collected magnets for other uses [19]. The ordinary method of REE recycling is based on hydrometallurgy using a strong acid, and the main materials are pure magnets and alloy scraps generated in a production facility. In reviewing cases of recycling, Rhodia recycled samarium-cobalt magnet (SmCo) by a selective leaching or solvent extraction using oxalic acid, sodium sulfate, tri-n-butyl phosphate, or di-(2-ethylhexyl) phosphoric acid (D2EHPA), after strong acid dissolution [8]. Wakayama University developed the solvent extraction process using aqueous hydrochloric acid solution and organic solvent and chemical reaction technology using ammonium oxalate to recover pure Nd from waste automobiles and home appliances [20]. Sumitomo Metal Industries developed the dry oxidation–reduction process that obtained Nd ingot from bond/sintered magnets and scraps, Advanced Industrial Science and Technology (AIST) recovered high-purity Nd magnet alloy (≥95%) by heating (300°C) and pulverizing waste magnets, and the University of Tokyo, Institute of Industrial Science (IIS) selectively extracted Nd and Dy by using high temperature (1,000°C) and magnesium chloride [19,21]. Hitachi recycled the REE magnets in household electric appliances by using processes of automated separation and dry extraction [3]. The Korea Institute of Geoscience and Mineral Resources (KIGAM) developed the hydrometallurgical recycling process to recover high-purity (3N) Nd and Dy from waste permanent magnets, and 60 tons of the scraps are being recycled annually at a pilot scale. KIRAM developed a dry extraction process (ReMat process) that can extract high-purity Nd and Dy from waste magnets by using the low melting point of magnesium [9].

In the fields of phosphors, owing to the importance of REMs, recycling research on phosphor in lighting equipment is actively performed in advanced countries, such as Japan and the United States. However, not only because of the low recovery rate of phosphors, but also the low quality of collected phosphors due to the inflow of other fluorescent substances and impurities, the recovery of high-purity REEs is still a challenge for practical use [22]. In reviewing cases of recycling, Rhodia/Solvay developed recycling technology that reuses recovered REEs for a new phosphor by pyrometallurgy and hydrometallurgy, after extracting and concentrating REEs from electric lights [18]. Japan is recently developing novel recycling technology that recovers REMs by acid leaching, after vitrification by adding oxides to the display phosphors [22]. In addition, Japan Oil, Gas and Metals National Corporation (JOGMEC) has developed the technologies of recycling oxides of Y, Eu, and Tb from waste phosphors, and extracting REMs from waste fluorescent lamps [23].

In the fields of batteries, various hydrometallurgical methods have been studied for recovering REEs used in the Ni-HM. In reviewing cases of recycling, Umicore and Rhodia recycled 7,000 tons of Ni-HM batteries in Belgium, by using the recycling process based on Umicore's patented ultra-high temperature smelting technology. Concentrated REEs after recovery are routinely sent to the Rhodia separation factory to produce REOs [8]. Honda Motor and Japan Metals & Chemicals extracted REEs from the waste Ni-HM batteries collected in Japan and obtained the REOs or REMs by using hydrometallurgy and molten salt electrolysis. JOGMEC studied methods for recycling lanthanum (La) and Ce, and Germany developed the pyrometallurgical process recovering REMs from slags generated during the thermal decomposition process of Ni-HM batteries. China studied the hydrometallurgical process using sulfuric acid to recover nickel, cobalt, and REMs [La, Ce, Nd, praseodymium (Pr)], and its recovery rate was about 95% at a laboratory level [16].

In other fields, display field (polishing) has come to the fore because after the use of Ce oxide slurry for chemical mechanical planarization, waste slurry can be relatively easily collected, and the purity of the recovered Ce oxide is also high. In reviewing cases of recycling, JOGMEC developed the novel technology that recycles Ce oxide abrasive by combining condensation and dispersion technologies of the waste slurry without acid leaching–based processes, and eight facilities (Fukushima University, etc.) in Japan also developed novel recycling technology that is free of chemical processes through an industry-university-research institution collaboration. China conducted research that recovers Sc and other REOs by leaching red mud, which occurs by the Bayer process in aluminum production, with hydrochloric acid, and Orbite Aluminae received a patent on technology recovering REEs from red mud [19,24,25].

### 3.2. *Hazard/risk of REEs recycling*

Significant caution may be required in the REE recycling processes because hazardous chemicals, which should be treated carefully, are used, or various harmful substances can be released or produced. The United Nations Environment Programme (UNEP) classified hazardous substance emissions in the REE recycling into 3 categories [26]. Primary emissions include lead, mercury, arsenic, polychlorinated biphenyls, sulfides, and ozone-depleting substances that may be contained in e-wastes, whereas secondary emissions include dioxins, furans, etc. that result from hazardous reaction due to inappropriate treatment of plastics with halogenated flame retardant. Tertiary emissions include harmful substances and reagents such as cyanide and leaching agents that are routinely used during recycling but are exposed outside by inappropriate management. This is becoming a significant problem in developing countries having small-scale and inappropriate recycling systems.

The hazards/risks for the main recycling processes are summarized as follows. First, in the collection process, harmful substances can be exposed to humans through inhalation, ingestion, and skin contact by leaking into the atmosphere, owing to breakage while collecting waste products. Second, volatile organic compounds (VOCs) and dust can be generated during the dismantling and separation processes, and the generated dust may contain various metal substances, including REEs. In the case of waste electronic products using refrigerants, such as refrigerators and air conditioners, Freon gas (chlorofluorocarbons, which are ozone-depleting substances), can be released into the atmosphere by an improper separation process. Because the LCD monitor contains mercury and other harmful metals, and the circuit board in electronic equipment contains flame retardants containing a synthetic resin and leads in the solder, they must be carefully dismantled. Mechanical dismantling and crushing are more likely to generate harmful dust and other particulate matters than manual processes. Dioxin can be generated at high temperature when crushing materials containing a brominated flame retardant, and therefore, thorough control systems are necessary. Magnet scrap powders generated after the polishing/cutting processes contain a large amount of fine powders (1 μm or less), which can ignite violently, or explode in an air-dried condition. In addition, energy injection, such as fossil fuel, that affects the environment is required for machine works. Landfill-generated metals and organics after dismantling and separation can contaminate surrounding groundwater, surface water, and soil and can be indirectly exposed to human body. Third, in the processing, because various methods are used depending on the application, different industrial process pollutants can be generated, respectively. Pyrometallurgy is an energy-intensive process requiring high temperature and produces VOCs and dioxin during the smelting process. Metallothermic reduction uses a high temperature of 1,000°C or higher, and molten salt electrolysis generates toxic gases, such as chlorine, fluorine, and explosive hydrogen gas. The electroslag remelting process, which is similar to the extraction process of REEs from raw ores, generates large quantities of solid wastes. In hydrometallurgy, strong acids, such as nitric acid and sulfuric acid, are used to melt raw materials, large amounts of chemicals are used in complicated procedures, and large amounts of wastewater are generated. In

**Table 1**
Summary of toxicological information about the REEs

| Element (abbreviation) | | Acute toxicity ($LD_{50}$) | | Repeated toxicity (NOEL, NOAEL, LOAEL) | | Reference dose or concentration (RfD, RfC) | | Remarks |
|---|---|---|---|---|---|---|---|---|
| | | Oral (mg/kg) | Inhalation (mg/L, 4h) | Oral (mg/kg/day) | Inhalation ($mg/m^3$) | Oral (mg/kg/day) | Inhalation ($mg/m^3$) | |
| Scandium (Sc) | $ScCl_3$ | 755 (rats) | | | | | | Sc is regarded as nontoxic, but toxic information is insufficient. |
| Yttrium (Y) | Y | >2,000 (♀, rats) | | | | | | Inhalation of Y citrate ($C_6H_5O_7Y$) induced dyspnea and pulmonary edema, and inhalation of Y chloride ($YCl_3$) induced pleural effusions, pulmonary hyperemia, and liver edema in rats. Water-soluble Y compounds have mild toxicity. The data of carcinogenicity are available. |
| | $Y_2O_3$ | >5,000 (rats) | >5.09 (rats) | 1,000[†] (28∼54-day, rats) | >20.63[*] (30-day, dogs) | | | |
| | $Y(NO_3)_3$ | 1650 (rats) | | ≥1,000[†] (32∼44-day, rats) | | | | |
| | $YCl_3$ | | | | | 0.004 | | |
| | $YF_3$ | >2,000 (♀, rats) | | | | | | |
| Lanthanum (La) | $La_2O_3$ | >10,000 (rats) | ≥5.3 (rats) | ≥525[†] (♂),<br>≥799[†] (♀)<br>(90-day, rats) | 0.5[‡] (28-day, ♂, rats) | 0.02 | | Injection of La induced reduced blood pressure, hyperglycemia, hepatic alterations, and spleen degeneration in *in vivo* experiments. |
| | $La(NO_3)_3$ | 4,500 (rats) | | ≥1,000[†] (46∼49-day, rats) | | | | |
| | $La_3$ | 2,621 (rats) | | ≥738[†] (♂),<br>≥1,122[†] (♀)<br>(90-day, rats) | | 0.005 | | |
| | $La_2(CO_3)_3$ | | >5.928 (rats) | 974[†] (♂),<br>1,480[†] (♀)<br>(90-day, rats) | | 0.5 | | |
| | $La(OH)_3$ | >2,000 (♀, rats) | | | | | | |
| | $LaF_3$ | >2,000 (♀, rats) | >5.1 (rats) | 1,000[†] (4∼6-week, rats) | | | | |
| Cerium (Ce) | $CeO_2$ | >5,000 (rats) | >5.05 (rats) | 1,000[*] (28∼54-day, rats) | 50.5[‡] (13-week, rats) | | 0.0009 | Injection of large doses of Ce induced cardiovascular collapse, resulting in death in experimental animals. Ce nitrate was reported to cause chromosomal aberrations. Ce fires produce toxic fumes. |
| | $Ce(NO_3)_3$ | 4,200 (♀, rats) | | 110[*], 330[†] (47-day, rats) | | | | |
| | $Ce(NO_3)_4$ | >2,000 (♀, rats) | | | | | | |
| | $CeCl_3$ | 2,800 (rats) | | | | | | |
| | $Ce_2(CO_3)_3$ | >5,000 (rats) | | 150[*], 450[†] (28∼54-day, rats) | | | | |
| | $CeF_3$ | ≥2,000 (rats) | >5.53 (rats) | 1,000[*] (4∼6-week, rats) | | | | |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| Neodymium (Nd) | $Nd_2O_3$ | >5,000 (rats) | >4.98 (rats) | 300[*], 1,000[†] (♂)<br>300[†] (♀)<br>(4～6-week, rats) | 0.5[‡]<br>(28-day, ♂, rats) | | | Nd compounds have low to moderate toxicities, but their toxic information is limited. Nd dust and salts irritate the eyes and mucous membranes highly and the skin moderately. Nd oxide $(Nd_2O_3)$ was reported as mutagen. |
| | $Nd(NO_3)_3$ | 2,750 (♀, rats) | | | | | | |
| | $NdCl_3$ | 5,068 (rats),<br>5,250 (mice) | | 840[†] (♂), 950[†] (♀)<br>(90-day, rats) | | 0.8[§] | | |
| | $Nd_2(CO_3)_3$ | >2,000<br>(♀, rats) | | | | | | |
| | $Nd(OH)_3$ | >2,000<br>(♀, rats) | | | | | | |
| | $NdF_3$ | >5,000 (rats) | | | | | | |
| Promethium (Pm) | | | | | | | | Pm may affect the human bone tissue. |
| Samarium (Sm) | $Sm(NO_3)_3$ | 2,900 (♀, rats) | | | | 0.00004[§] | | Water-soluble Sm compounds are slightly toxic. |
| | $SmCl_3$ | 3,200 (rats) | | | | 0.9[§] | | |
| Europium (Eu) | $Eu_2O_3$ | | | | | 0.002 | | Eu was reported to be relatively nontoxic compared to other heavy metals. |
| | $Eu(NO_3)_3$ | >5,000<br>(♀, rats) | | | | | | |
| | $EuCl_3$ | 5,000 (mice) | | 200[*]<br>(28-day, rats) | | 0.03 | | |
| Gadolinium (Gd) | Gd | >2,000<br>(♀, rats) | | | | | | Free Gd ions are highly toxic, and mutagenicity data (micronucleus, chromosomal effects) is available. About 0.03～0.1% of anaphylactoid reactions were reported. |
| | $Gd_2O_3$ | >1,000<br>(♀, rats) | >5.04 (rats) | 1,000[†]<br>(11～12-week, rats) | | | 0.002 | |
| | $Gd(NO_3)_3$ | >5,000<br>(♀, rats) | | | | | | |
| Thulium (Tm) | $TmCl_3$ | 6,250 (mice) | | | | | | Water-soluble Tm compounds are slightly toxic. |
| Dysprosium (Dy) | $Dy_2O_3$ | >5,000 (rats) | | | | | | Water-soluble Dy compounds such as Dy chloride and Dy nitrate are mildly toxic. |
| | $Dy(NO_3)_3$ | 3,100 (♀, rats) | | | | | | |
| | $DyCl_3$ | 7,650 (mice) | | | | | | |
| Ytterbium (Yb) | $Yb_2O_3$ | >2,000<br>(♀, rats) | >1.31 (rats) | 1,000[†]<br>(15～16-week, rats) | | | | All Yb compounds are regarded as highly toxic due to their irritation for the skin and eye and the possibility of teratogenic effects. |
| | $Yb(NO_3)_3$ | 3,100 (♀, rats) | | | | | | |
| | $YbCl_3$ | 6,700 (mice) | | | | | | |
| | $YbF_3$ | >2,000 (♀, rats) | | | | | | |
| Lutetium (Lu) | $Lu_2O_3$ | >2,000<br>(♀, rats) | | ≥1,000[†]<br>(12～13-week, rats) | | | | Inhalation of Lu fluoride and oxide powders is dangerous. Water-soluble Lu compounds are mildly toxic. |
| | $Lu(NO_3)_3$ | >300, ≤2,000<br>(♀, rats) | | | | | | |
| | $LuCl_3$ | 7,100 (mice) | | | | 0.5[§] | | |
| Praseodymium (Pr) | $Pr_2O_3$ | >2,000 (rats) | >5.21 (rats) | 1,000[†]<br>(4～6-week, rats) | | | | Pr compounds have low to moderate toxicities. |
| | $Pr(NO_3)_3$ | 3,500 (♀, rats) | | | | | | |
| | $PrCl_3$ | 4,500 (♂, mice) | | ≥10,000[†] mg/kg diet<br>(90-day, rats) | | 0.8[§] | | |
| | $Pr_2(CO_3)_3$ | >2,000 (♀, rats) | >5.25 (rats) | | | | | |

(*continued on next page*)

**Table 1** (*continued*)

| Element (abbreviation) | | Acute toxicity ($LD_{50}$) | | Repeated toxicity (NOEL, NOAEL, LOAEL) | | Reference dose or concentration (RfD, RfC) | | Remarks |
|---|---|---|---|---|---|---|---|---|
| | | Oral (mg/kg) | Inhalation (mg/L, 4h) | Oral (mg/kg/day) | Inhalation ($mg/m^3$) | Oral (mg/kg/day) | Inhalation ($mg/m^3$) | |
| Terbium (Tb) | $Tb(NO_3)_3$ | >5,000 (♀, rats) | | | | | | Tb compounds have low to moderate toxicities, but their toxicity information is limited. |
| | $TbCl_3$ | 5,100 (mice) | | | | | | |
| Holmium (Ho) | $Ho(NO_3)_3$ | 3,000 (rats) | | | | | | Ho compounds have low acute toxicity. |
| | $HoCl_3$ | 7,200 (mice) | | | | | | |
| Erbium (Er) | $Er_2O_3$ | >2,000 (rats) | >5.09 (rats) | 1,000[†] (♂),<br>300[*] (♀)<br>(4~6-week, rats) | | | | Er compounds have low to moderate toxicities, but their toxicity information is limited. |
| | $ErCl_3$ | 6,200 (mice) | | | | | | |

$LD_{50}$, lethal dose 50; NOEL, no-observed-effect level or concentration; NOAEL, no-observed-adverse-effect level or concentration; LOAEL, lowest observed adverse effect level or concentration.

[*] NOEL value.

[†] NOAEL value.

[‡] LOAEL value.

[§] Provisional subchronic RfD.

particular, nitrogen oxides and chlorine gas are generated owing to the use of nitric acid or aqua regia (mixture of concentrated nitric acid and hydrochloric acid) during leaching. Furthermore, as solvent extraction, leaching and selective precipitation methods are similar to the extraction of REEs from raw ores, where harmful slag, chemicals, and particulate matters can occur. Versatile gas-phase extraction used in magnet recycling uses a large amount of chlorine gas and generates strongly corrosive aluminum chloride. Because recycling of mine tailings is performed by using existing processes and devices in the mining field, process-generated contaminants are considered as similar to those that occur where raw ores are processed [3,8,9,16,26].

Reviewing issues and hazard/risk of ore processing is necessary because some recycling methods are similar to that of processing, which routinely uses a hydrometallurgical process that produces a large amount of harmful substances [3]. According to the Hurst report, about 8.5 kg of fluorine and 13 kg of dusts are generated to produce 1 ton of REEs, and the use of concentrated sulfuric acid (96%) during the high-temperature calcination process causes the production of 9,600 to 12,000 $m^3$ of waste gas containing hydrofluoric acid, sulfur dioxide, and concentrated dust, and 75 $m^3$ of acidic wastewater [27]. In addition, the use of the saponification extraction method, which has until recently been used in China, produced 20,000 to 25,000 tons of wastewater, whose total concentration of ammonia-nitrogen was measured as 300 to 5,000 mg/L in China as of 2015 [16]. A significant environmental issue, showing that the total dissolved solids concentration of groundwater near the Mountain Pass mine of Molycorp increased from (360–800) to approximately 10,000 mg/L, has arisen before, in the United States, owing to the application of onsite percolation-type surface impoundments that use sodium hydroxide to neutralize hydrochloric acid in the wastewater [3]. Four waste streams in ore processing were evaluated as hazardous by the United States Environmental Protection Agency (US EPA); (1) waste solvent (ignitability), (2) solvent extraction crud (ignitability), (3) spent lead filter cake (toxicity), and (4) waste zinc contaminated with mercury (toxicity) [28].

### 3.3. *Toxicity and occupational safety and health issues of REEs*

As the use of REEs has increased, concern for industrial health has been gradually increasing, and the reports on bioaccumulation, epidemiological survey, *in vivo* and *in vitro* toxicity studies, and related issues for REEs have been limited up to now [12,29].

In the toxicity studies, REE compounds showed low acute toxicities, because most of their oral and inhalation lethal dose 50 ($LD_{50}$) in experimental animals were higher than about 2,000 mg/kg or 5,000 $mg/m^3$ (4 h) [30,31]. On the other hand, the lowest observed adverse effect level was 50.5 $mg/m^3$ due to increased alveolar epithelial hyperplasia in the 13-week inhalation toxicity study of Ce oxide in rats, suggesting a reference concentration (RfC) of 0.0009 $mg/m^3$ [3,30]. Recently, the lowest observed adverse effect level of La and Nd oxides was determined as 0.5 $mg/m^3$ in rats, owing to inflammatory changes and pulmonary alveolar proteinosis in the lungs in the 4-week inhalation toxicity studies. In *in vitro* studies using macrophages, Ce and Nd showed a probability of pulmonary toxicity and potential fibrosis [32]. In a 30-day repeated oral administration of Ce chloride in mice, liver toxicity was observed to be 10 and 20 mg/kg showing histopathological changes, such as congestion, fatty degeneration, and cloudy swelling, with increased alanine aminotransferase [33]. Dy and Nd chlorides induced conjunctivitis in the Draize eye irritation test using rabbits, and severe irritation on the scratched skin of rabbits in the skin sensitization test [34,35]. Although there have been a few research results concerning carcinogenicity and mutagenicity,

**Table 2**
Summary of worker health, safety, and working environment management practices

| Area | Management practices | Details |
|---|---|---|
| Working process and environment | Process automation | • Minimizing human exposure time to chemicals |
| | Sealing and isolation of dust-generating processes | • Minimizing the size of the opening as much as possible<br>• Installing a scattering preventing apparatus such as a hopper |
| | General and local ventilation installation | • Ensuring a suitable local ventilation in working area<br>- installing in the following order: hood (closing type if possible); duct; air cleaning device; blower; exhaust vents<br>- maintaining the velocity control of local exhaust based on the Safety and Health Regulation (No. 429)<br>• Installing a general ventilation for negative pressure maintenance if having a difficulty in installing a local ventilation |
| | Work environment monitoring | • Collecting and analyzing the samples in working area to check the standard excess (8-hour TWAs) |
| | Protective equipment wearing | • Checking mask fitting, suitability, and certification |
| | Separate storage of combustible materials | • Avoiding ignition sources |
| Health monitoring | Medical diagnosis | • Performing before and after work including an occasional examination<br>• Needing differentiation from other similar diseases |
| Health and safety education | Regular education based on the MSDS and technical instruction | • Understanding material toxicity and physiochemical properties, standard operation knowhow, correct use of equipment and device, and emergency treatment |

MSDS, material safety data sheet; TWA, time-weighted average.

the US EPA has not yet classified REEs as carcinogens [56]. Table 1 summarizes the overall toxicological information for REEs [12,30,31,36–38].

In the occupational safety and health issues, short-term inhalation exposure of REEs induced acute irritant bronchitis, while long-term inhalation exposure of REEs induced pneumoconiosis and progressive pulmonary fibrosis. However, it has been controversial as to whether the progression of interstitial pulmonary fibrosis is due to REEs, or due to radioactive contaminants. This was because it is possible that stable REE compounds induce a benign pneumoconiosis by foreign body reaction, but its progress into an interstitial fibrosis after termination of exposure is uncertain [39–42]. Cases of fingertip injury due to Nd's strong magnetic force and biocompatibility issues of Nd on medical uses were reported. Moreover, intravenous injection of 3–18 mg of Nd compounds, such as Nd nitrate, Nd lactate, and Nd acetate, caused fever, chills, myalgia, spastic abdominal pain, hemoglobinemia, and hemoglobinuria, and prolonged blood clotting time by 2- to 4-fold. Nd dust can cause lung embolism and liver damage in exposed workers. Y, Eu, Dy, erbium (Er), and ytterbium (Yb) are dangerous because their dust forms can be combusted or exploded in air. In particular, Dy can be combusted by sparks or static electricity, even when existing in the form of a thin foil, and when reacting with water, generates flammable and explosive hydrogen gas. In addition, the fire that occurs by Dy cannot be extinguished by water. Exposure of Ce can cause itching, skin lesions, and sensitivity to heat in workers, owing to its irritancy. While Ce is a strong reductant and can be combusted in air at 65–80°C, $CeO_2$ is a strong oxidizer at high temperatures, and easily reacts with combustible organic materials. Lutetium nitrate [$Lu(NO_3)_3$] can be burned or exploded by heating. Treatment of Sc needs special attention in the workplace (metal halide lamp) because its gas form mixed with other metals or compounds can be exposed to workers. Exposure of Y compounds can cause coughing, shortness of breath, cyanosis, and chest pain in workers. REE oxides and fluorides, including La, have induced pneumoconiosis before among workers treating carbon arc lamps. Lu was reported to be accumulated in the bones (much), liver, and kidneys, although there was no information for the biological effects. Holmium (Ho) and thulium (Tm) compounds are known to stimulate the metabolism of the human body, but their biological effects are still controversial [12,29,43]. Dermal fibrosis (nephrogenic systemic fibrosis) was reported by gadolinium (Gd) used as contrast agents for magnetic resonance images [44]. A recent study reported that Gd contrast agent used in magnetic resonance imaging has the possibility of causing severe kidney disease by passing through the blood-brain barrier and accumulating in the brain tissue, and the distribution and chemical behavior of Gd in the brain were investigated [45].

In the epidemiological surveys, serum protein, albumin, β-globulin, alanine aminotransferase, triglyceride, and IgM significantly decreased, and blood cholesterol significantly increased in the blood examination for residents in Jiangxi Province (southern China), where REE exposure has frequently occurred [46]. In other studies conducted in the same area, the intelligence quotient of children was significantly lower than that in the control area, and the incidence of arteriosclerosis in adults was also high [47]. In another case-control study, correlation between Ce exposure and early acute myocardial infarction for elderly people aged about 70 years in Europe and Israel was analyzed. In the results of 684 cases and 724 controls, the average Ce levels in toenail samples were

186 and 173 μg/kg, respectively, and the gap between them was widened, after correcting for age, residence, and cardiovascular disease risk factors, indicating confirmation of the clear relationship between the two factors [48].

### *3.4. Requirements for worker safety in the REE recycling*

A verified monitoring system for the REE recycling process is required to dispel social worries, such as potential human toxicity and environmental pollution, that can occur in the complicated circular economy. In particular, Ali [49] has previously referred to the importance of management for radiation exposure, health, environmental impacts, and safety in the REEs industry. However, radiation exposure is a major problem in ore processing and is not directly related to recycling. Concern for occupational health has focused on human exposure to radioactive substances during ore processing, and related regulatory policies were reported in the United States, Canada, China, etc. On the other hand, the influence of REMs or REE compounds on the human body had only been reported sporadically in case reports, and it came to be examined and reviewed systematically from the early 1990s, and then problematized by Hirano and Suzuki [50]. To date, regulation for REEs was established for Y and Ce, only. The National Institute of Occupational Safety & Health (NIOSH) recommended the time-weight average and immediately dangerous to life or health of Y compounds as 1 and 500 mg/m$^3$, respectively, and Russia regulated the occupational exposure limit of Ce oxide as 5 mg/m$^3$ [12]. Thus, the exposure limit for overall REEs is required to be established by performing comprehensive hazard/risk assessment and to be complemented by checking exposure conditions in the workplace through an onsite field survey. In addition, occupational diseases should be prevented by periodic monitoring of workers' health [29]. Environmental monitoring for air, water, and soil requires a thorough wastewater treatment system, including pH monitoring, as with other large-scale factories using complex organic or inorganic reagents, and failure to monitor leads to secondary contamination due to environmental exposure. Most of the work-polluting environments occurred during ore processing (smelting), and similar methodology is used in the recycling process. REE carbonate provides a natural buffer to alleviate acidosis resulting from various acid leaching, but inversely, excessive carbonate can cause alkalosis. A case was reported of wastewater being leaked due to damage of a buried pipeline in a mining facility belonging to Molycorp in Mountain Pass [3]. To ensure safety in the workplace, thorough management of hazardous substance, appropriate protective equipment, and regular safety education are necessary in the leaching process of the REE smelting and recycling because nitrogen oxides or chlorine gas can occur by using nitric acid or aqua regia during the process. For better monitoring system, it is necessary to refer to the relevant regulation, standard, or legislation in other industries dealing with solvent extraction, electrolytic process, and high-functioning chemicals. For example, although the REE processing facility belonging to Rhodia in La Rochelle, France, is located near a small town, unfavorable cases, such as the resistance of residents or serious safety accidents, have not been reported so far because the facility was registered as the Installations Classified for the Protection of the Environment and operated in accordance with strict safety standards. The monitoring system in that facility may be a blueprint for other REE processing facilities [45]. We suggest the practices for worker health, safety, and working environment management in Table 2.

Currently applied processes are required to be additionally developed to reduce the risks to human health and the environment. There are chemical, biological, biochemical, biological adsorption, and physicochemical methods for removing REEs in wastewater, and REEs are routinely recovered by acid leaching and selective precipitation. Recently, additional research studies have been developed based on solid-solid extraction, ultrasonic extraction, supercritical carbon dioxide, plasma separation technologies, and membrane technology to treat trace amounts of REEs more effectively. Adsorption-based technology, such as acid leaching, is currently used for recovering REEs from mine wastes and environmental samples; but the use of electrochemical, membrane filtration, oxidation, photocatalyst, biological treatment, or a combination thereof is necessary for the efficient recovery of REEs in the future [45]. Innovative recycling research has been actively conducted in Japan, and a low-cost, high-efficiency, environment-friendly technology was reported, which used salmon milt containing phosphate that has high adsorption affinity for REEs [51]. The Comet Traitements of Belgian company also developed bio-hydrometallurgical technology with Liège University to recover valuable metals from metal scraps. The technology used bacteria regenerating iron (III) (oxidants) in sulfuric acid solutions and is under further research for REM recovery [52].

For better occupational safety and health, policy support and infrastructure construction are essential. Looking at the related regulatory policies in the REEs industry (ore processing) due to environmental problems, the Ministry of Environmental Protection of China (MEP) issued the "Emission Standards of Pollutants from Rare Earths Industry" for environmentally friendly development in 2011. This standard compelled the reduction of ammonia nitrogen per 1 L of sewage and wastewater from (300–5,000) to 25 mg to prevent serious environmental pollution by the large quantities of acids, alkalis, and extractants used in the REE smelting and separation from 2012 to 2013, and to reduce to 15 mg afterward [53]. The Ministry of Industry and Information Technology of China (MIIT) also issued the "Entry Criterions for Rare Earth Industry" in 2010. This document defined minimum standards concerning the production capacity, capital ratio for fixed assets, used techniques and equipments, and REE recovery rates, whereby related facilities were obliged to be fully equipped with wastewater, waste gas, and solid waste treatment systems, and to dispose fluorine-containing solid wastes separately. In addition, the use of harmful techniques, such as the ammonia saponification process, and electrolyzing metals by their chlorides, was prohibited. The standards for ore dressing recovery rate of mixed REEs, including bastnasite and ion-adsorption type of REEs, were more than 72% and 70%, respectively; the standards for wastewater recycling rate during these processes were more than 85% and 90%, respectively; and the purity standard for refined REMs was more than 92%. The United States planned protection measures include a series of treatments, such as the recovery of salts (hydrochloric acid, sodium hydroxide, sodium hypochlorite), reuse of water, and reduction of water usage during the ore dressing process, collection, and concentration of leachate derived from ore stockpiles, groundwater remediation systems, flue gas treatment plants, separate disposal on exterior landfills for lead-containing hazardous wastes, and area remediation after mining, to prepare for reopening at Mountain Pass in California [16]. In terms of worker safety, the US EPA has announced the risk guidance document that the source and mechanism for chemical release, transport or retention medium, potential human exposure point and affected medium, and exposure route (oral, dermal, inhalation) at the point must be included in a completed exposure pathway [54].

## 4. Discussion

The REEs are widely used in various industrial fields, such as electronics, petrochemistry, metallurgy, military facilities, and medicine, owing to their specific physiochemical properties, and their usage is expected to increase as essential industrial materials in the new growth engine of the 21st century despite the small proportion in the products. However, demanding countries have prepared measures such as the development of new mines, reopening of closed mines, and recycling and substitution of resources, owing to localized deposits of REEs, environmental pollution, and political means. In particular, recycling is recommended from the viewpoint of the “balance problem” of resources, as well as the efficiency of resource circulation and absence of radiation exposure fatal to the human body, compared to ore processing. This means that the production of unnecessary resources (La and Ce), which is caused by the difference of individual element content in the REE ores, is avoided by recycling a Nd having a relatively low content in the ores and has attracted wide interest in China [55]. Japan is now becoming an advanced recycling country by activating urban mining and political support, and in keeping with global trends, resource-poor Korea also joined ISO/TC 298 and is promoting a REE recycling industry.

REE recycling is globally applied to the fields of magnets, abrasives, phosphors, and batteries, and there are collection, dismantling/separation, pyrometallurgy, hydrometallurgy, electrochemical process, and materialization technology in the processes. Despite various research efforts, there are only a few practical cases, owing to the lack of government support and infrastructure for a REEs recycling industry, with inefficient collection, technical problems, and deficient incentives. The UNEP reported that the practical recycling rate of REEs was less than 1% in 2011 [56]. However, Yale University in the United States suggested that Ce, La, Nd, and Y, whose production is over 85% in the global production of REEs, can be recycled, although there are technical challenges [57]. Moreover, 20%–30% of REE magnets are discarded during the manufacturing process, and the 10–20 g of Nd–Iron (Fe)–Boron (B) magnets in each hard disk drive were reported to be equal to 6,000–12,000 tons of Nd–F–B alloy [16,58]. Therefore, producing countries, as well as demanding countries, have performed continuous research for practical REE recycling applications, to avoid enormous resource loss. KIGAM developed its own hydrometallurgical process and recycled 60 tons of waste magnets annually on a pilot scale through a subsidiary company in Korea.

While the importance of REE recycling has been emphasized, appropriate management measures are essential because harmful substances to the environment and human bodies can be handled during the recycling processes. On the basis of the UNEP classification category, collection, dismantling, and separation can cause primary emissions. Because these processes mainly need physical operation, and electronic devices contain mercury, lead, arsenic, polychlorinated biphenyls, VOCs, and Freon gas, harmful substances or REEs containing dusts can be exposed through dermal, oral, or inhalational pathways by the manual and mechanical dismantling or breakage at the time of collection. It is still an important concern for the complete recovery of mercury during lamp phosphor recycling [19]. In addition, pulverized fine powders of magnet scrap can explode in air, and in the separation step, dioxin can occur by high temperature when crushing brominated flame retardant containing items. In the processing step, the use of recycling methods similar to ore processing is considered to cause more potential impacts on human and environment. In particular, in hydrometallurgy, which has been widely used not only in Korea, but also worldwide as a classical method, a large amount of chemicals, such as leaching agent and strong acids, are used, and so lots of wastewater, and harmful nitrogen oxides and chlorine gas, can be generated (tertiary emissions). Pyrometallurgy using high temperature can also cause harmful gases, such as chlorine, fluorine, VOCs, and dioxin and solid wastes. According to the UNEP report, 50% of waste products for recycling were illegally exported to Africa or Asia and recycled indiscriminately outdoors without any protective equipment and consequently became a major threat to workers and the environment. For example, incineration or inappropriate smelting of halogenated flame retardants containing plastics can cause dioxins or furans (secondary emissions). Fatal injury including fatality occurred in about 4,600–5,200 cases from 2011 to 2016, and among them, the accidents caused by hazardous substances and harmful environmental exposure, except for electric shock accident, gradually increased every year by about 3.3%–6.1% [59]. REEs were also reported to induce disorders of respiratory, neurological, cardiovascular systems in epidemiological surveys, and consistent with these results, damage of the lung (inhalation toxicity) and liver (oral toxicity) was reported, including skin irritation, physical injury, and risk of ignition and explosion in powder form in toxicity studies and occupational safety and health issues. However, further studies are required for accurate hazard/risk assessment, because the epidemiological surveys mostly focused on the Chinese, and reflected the data of multiple REEs exposure, and long-term inhalation toxicity study over 3 months has not yet been performed, except for Ce [11].

Monitoring systems or guidelines regarding health, environmental impacts, and the safety of applied processes are essential to prevent industrial accidents and environmental pollution in the future recycling industry. The overall exposure limit for REEs should be established and verified by performing additional investigation of REE toxicity, systematic epidemiological survey, and onsite field survey. If REE exposure is suspected through periodic health monitoring, quick outpatient clinic should be carried out with appropriate treatment. Monitoring systems are also considered to be established with reference to the guidelines and hazard/risk assessment models in the relevant work fields because processing methods, such as hydrometallurgy and electrolytic refining, can be used universally in the overall mineral processing that includes REEs [3]. Gwenzi et al. [45] reported the conceptual framework in the REEs industry, which consists of 6 consecutive steps for the assessment and mitigation of human and ecological health risks: “hazard identification”, “risk analysis”, “risk evaluation”, “evaluation of interventions”, “implementation of interventions”, and “monitoring and evaluation”. In addition, various health impacts assessment (HIA) process models and HIA good practice guidelines have been developed, which focus on regional, national, and international levels, depending on the literature review of the International Council on Mining and Metals (ICMM). HIA consists of two categories of Occupational HIA (OHIA), which deals with physical injury in the workplace, such as accidents, hearing loss, musculoskeletal disorder, sunlight and heat exposure, and radiation; and Community HIA (CHIA), which deals with potential human health risks due to contaminated surrounding environments, such as nutritional disorders, physical injury, chronic diseases, and mental health [60].

Meanwhile, the field of occupational safety and health is easily neglected in the country where the REs recycling industry is in the initial stage of development, because of low level of awareness and poor surroundings for REE recycling. Thus, government support, infrastructure construction, and technical development for high-efficiency, low-cost, environmentally friendly methods by government-funded research institutes and related companies are required, to build both REE recycling industrial development, and advanced occupational safety and health conditions. Reduction of chemicals used in the original processes prevents the generation of harmful substances, and the improvement of wastewater/wastes treatment technology helps to effectively collect trace amounts of REEs and pollutants, resulting in completely blocking the human and ecosystems exposure. Two beneficial effects of increased recovery rate and reduced contaminants can be expected by developing an innovative and effective recycling technology using living organisms or new materials. In addition, it is possible to provide overall information about all human exposure routes indirectly by developing *in vivo* analysis and biomarkers for REEs that are used in human risk assessment.

## 5. Conclusion

It is necessary to establish verified monitoring systems or guidelines in the workplace that focus on health, environmental impacts, and safety, to protect worker health and environments in the future recycling industry. For health, systematic onsite field and epidemiological surveys, and long-term exposure study of laboratory animals, are required for the identification of hazardous level and establishment of regulatory level in the workplace, and occupational diseases should be prevented in advance by periodic health monitoring. For environmental impacts and safety, secondary damage through environmental media should be prevented by thorough wastes management, and the wearing of protective equipment and adequate safety education are needed, to prevent occupational accidents, such as fire, explosion, physical injury, and exposure of harmful substances. Because the REE recycling processes are similar to those applied to other heavy metals, it is recommended to actively use the well-established guidelines and hazard/risk assessment models in related fields. In addition, it is necessary to develop innovative and environmentally friendly recycling technologies, *in vivo* analytical methods, and biomarkers. In particular, government support is essential in the country where the REEs recycling industry is in the initial stage of development, and through these efforts, the occupational safety and health status will be improved, along with the establishment of advanced REE recycling industry.

## Conflicts of interest

The authors declare that there are no conflicts of interest.

## Acknowledgments

This study was supported by the intramural research fund of the Occupational Safety and Health Research Institute (OSHRI).

## References

[1] Humphries M. Rare earth elements: the global supply chain. Washington, DC (USA): Congressional Research Service; 2013. 27 p. Report No.: R41347.

[2] Tong SL, Zhu WZ, Gao ZH, Meng YX, Peng RL, Lu GC. Distribution characteristics of rare earth elements in children's scalp hair from a rare earths mining area in southern China. J Environ Sci Health A Tox Hazard Subst Environ Eng 2004;39:2517–32.

[3] US EPA (United States Environmental Protection Agency). Rare earth elements: a review of production, processing, recycling, and associated environmental issues; 2012. Washington, DC (USA), Report No.: EPA 600/R-12/572.

[4] Haque N, Hughes A, Lim S, Vernon C. Rare earth elements: overview of mining, mineralogy, uses, sustainability and environmental impact. Resources 2014;3(4):614–35.

[5] Abbas SF, lee SH, Lee B, Kim BS, Kim TS. Trends and implications of international standardization for rare earths. J Korean Powder Metall Inst 2018;25(2):165–9.

[6] European Commission. Critical raw materials for the EU – report of the Ad-hoc Working Group on defining critical raw materials; 2010. 85 p.

[7] US DOE (United States Department of Energy). Critical materials strategy; 2012. 196 p. Washington, DC (USA), Report No.: DOE/PI-0009.

[8] Binnemans K, Jones PT, Blanpain B, Gerven TV, Yang Y, Walton A, Buchert M. Recycling of rare earths: a critical review. J Clean Prod 2013;51:1–22.

[9] Cho JH, Lee HS, Kim CH, Shin KH. Secondary material flow analysis to activate recycling of rare earth magnet. Seoul (Korea): Korea Environment Institute; 2013. 136 p. Report No.: 978–89–8464–772–5 93530.

[10] Pagano G, Guida M, Tommasi F, Oral R. Health effects and toxicity mechanisms of rare earth elements – knowledge gaps and research prospects. Ecotoxicol Environ Saf 2015;115C:40–8.

[11] Pagano G, Aliberti F, Guida M, Oral R, Siciliano A, Trifuoggi M, Tommasi F. Human exposures to rare earth elements: state of art and research priorities. Environ Res 2015;142:215–20.

[12] Rim KT, Koo KH, Park JS. Toxicological evaluations of rare earths and their health impacts to workers: a literature review. Saf Health Work 2013;4:12–26.

[13] Waring PM, Watling RJ. Rare earth deposits in a deceased movie projectionist. A new case of rare earth pneumoconiosis? Med J Aust 1990;153:726–30.

[14] Dillow CA. New international project aims to track U.S. electronic waste for recycling [Internet]; 2011 [cited 2019 Mar 27]. Available from: https://www.popsci.com/science/article/2011-05/new-international-project-aims-track-us-electronic-waste-recycling.

[15] Clenfield J, Shiraki M. Hitachi develops machine to recycle rare earths to cut reliance on China [internet]; 2010 [cited 2019 Mar 27]. Available from: http://www.hitachi.com/New/cnews/101206.html.

[16] Schüler D, Buchert M, Liu R, Dittrich S, Merz C. Study on rare earths and their recycling. Freiburg (Germany): Öko-Institut e.V.; 2011. 140 p.

[17] Gambogi J. Mineral commodity summaries 2015. Reston (USA): U.S. Geological Survey; 2015. 196 p.

[18] Tsamis A, Coyne M. Recovery of rare earths from electronic wastes: an opportunity for high-tech SMEs. Brussels (Belgium): European Parliament; 2015. 42 p. Report No.: IP/A/ITRE/2014-09.

[19] Park JY. Recycling technology and industrial trends of waste rare earths. Seoul (Korea): Korea Environmental Industry and Technology Institute; 2017. 10 p [in Korean].

[20] Jakobsson LK, Kennedy MW, Aune RE, Tranell G. Recovery of rare earth elements from the ferrous fraction of electronic waste. In: Randolph EK, Bart B, Christina M, Elsa O, Diran A, John H, Anne K, Brajendra M, Neale RN, Jeff S, editors. Rewas 2016: towards materials resource sustainability. New York (USA): Springer; 2016. p. 89–93.

[21] Shirayama S, Okabe T. Selective extraction of Nd and Dy from rare earth magnet scrap into molten salt. In: Mishra B, Fuma A, Bhandhubanyong P, editors. Minerals, metals and materials society–3rd international conference on processing materials for properties 2008. New York (USA): Curran; 2009. p. 469–74.

[22] Zhang K, Kleit AN, Nieto A. An economics strategy for criticality–Application to rare earth element Yttrium in new lighting technology and its sustainable availability. Renew Sust Energ Rev 2017;77(C):899–915.

[23] Tan Q, Li J, Zeng X. Rare earth elements recovery from waste fluorescent lamps: a review. Crit Rev Environ Sci Technol 2015;45(7):749–76.

[24] King AH, Eggert RG, Gschneidner KA. The rare earths as critical materials. Handbook Phys Chem Rare Earths 2016;50:19–46.

[25] Wang H, Liang Y, Jiang P. Recovering the rare earth from red mud generation. Rare Earth Lett 2005;1008–5939. 10-1005-04.

[26] Schluep M, Hagelueken C, Kuehr R, Magalini F, Maurer C, Meskers C, Mueller E, Wang F. Recycling – from e-waste to resources. Berlin (Germany): United Nations Environment Programme & United Nations University; 2009. 94 p. Report No.: DTI/1192/PA.

[27] Hurst C. China's rare earth elements industry: what can the west learn? Potomac (USA). Institute for the Analysis of Global Security (IAGS); 2010. 42 p.

[28] US EPA (United States Environmental Protection Agency) [Internet]. In: Identification and description of mineral processing sectors and waste streams 1991 [cited 2019 Mar 27]. Available from: https://archive.epa.gov/epawaste/nonhaz/industrial/special/web/html/index-8.html; 1991.

[29] Shin SH, Rim KT, Kim JC. Evaluation of rare earths viewed from the occupational health. J Korean Soc Occup Environ Hyg 2016;28(1):1–17.

[30] ECHA (European Chemicals Agency). ECHA database on registered substances [Internet]; 2019 [cited 2019 Mar 27]. Available from: http://echa.europa.eu/information-on-chemicals/registered-substances.

[31] Damian P. Stand alone report 14 – a literature review of the health and ecological effects of the rare earth elements. Lakewood (USA): United States Nuclear Regulatory Commission; 2014. 47 p.

[32] Palmer RJ, Butenhoff JL, Stevens JB. Cytotoxicity of the rare earth metals cerium, lanthanum, and neodymium *in vitro*: comparisons with cadmium in a pulmonary macrophage primary culture system. Environ Res 1987;43(1):142–56.

[33] Cheng J, Cheng Z, Hu R, Cui Y, Cai J, Li N, Gui S, Sang X, Sun Q, Wang L, Hong F. Immune dysfunction and liver damage of mice following exposure to lanthanoids. Environ Toxicol 2014;29(1):64–73.

[34] Haley TJ, Koste L, Komesu N, Efros M, Upham HC. Pharmacology and toxicology of dysprosium, holmium and erbium chlorides. Toxicol Appl Pharmacol 1966;8:37–43.

[35] Haley TJ, Komesu N, Efros M, Koste L, Upham HC. Pharmacology and toxicology of praseodymium and neodymium chlorides. Toxicol Appl Pharmacol 1964;6:614–20.

[36] US EPA (United States Environmental Protection Agency). Provisional peer-reviewed toxicity values for stable (nonradioactive) neodymium chloride (CASRN 10024-93-8). Cincinnati (USA); 2009. 24 p.

[37] Kim YS, Lim CH, Shin SH, Kim JC. Twenty-eight-day repeated inhalation toxicity study of nano-sized neodymium oxide in male sprague-dawley rats. Toxicol Res 2017;33(3):239–53.

[38] Shin SH, Lim CH, Kim YS, Lee YH, Kim SH, Kim JC. Twenty-eight-day repeated inhalation toxicity study of nano-sized lanthanum oxide in male sprague-dawley rats. Environ Toxicol 2017;32(4):1226–40.

[39] Harbison RD. Hamilton & hardy's industrial toxicology. 5th rev. New York (USA): Mosby, Inc.; 1998. 682 p.

[40] Haley PJ. Pulmonary toxicity of stable and radioactive lanthanides. Health Phys 1991;61(6):809–20.

[41] Vocaturo G, Colombo F, Zanoni M, Rodi F, Sabbioni E, Pietra R. Human exposure to heavy metals. Rare earth pneumoconiosis in occupational workers. Chest 1983;83:780–3.

[42] US EPA (United States Environmental Protection Agency). Provisional peer-reviewed toxicity values for stable lutetium (CASRN 7439-94-3); 2018. 26 p. New York (USA); Report No.: EPA/690/R-18/003.

[43] Beaser SB, Segel A, Vandam L. The anticoagulant effects in rabbits and man of the intravenous injection of salts of the rare earths. J Clin Invest 1942;21(4): 447–54.

[44] Ramalho J, Semelka RC, Ramalho M, Nunes RH, AlObaidy M, Castillo M. Gadolinium-based contrast agent accumulation and toxicity: an update. AJNR Am J Neuroradiol 2016;37(7):1192–8.

[45] Gwenzi W, Mangori L, Danha C, Chaukura N, Dunjana N, Sanganyado E. Sources, behaviour, and environmental and human health risks of high-technology rare earth elements as emerging contaminants. Sci Total Environ 2018;636:299–313.

[46] Zhang H, Feng J, Zhu W, Liu C, Xu S, Shao P, Wu D, Yang W, Gu J. Chronic toxicity of rare-earth elements on human beings: implications of blood biochemical indices in REE-high regions, south Jiangxi. Biol Trace Elem Res 1999;73:1–17.

[47] Zhang H, Zhu WF, Feng J. Subchronic toxicity of rare earth elements and estimated daily intake allowance. In: Ninth annual V.M. Goldschmidt conference [internet] 1999 [cited 2019 Mar 27];1:7025. Available from: https://www.lpi.usra.edu/meetings/gold99/pdf/7025.pdf; 1999.

[48] Gómez-Aracena J, Riemersma RA, Gutiérrez-Bedmar M, Bode P, Kark JD, Garcia-Rodríguez A, Gorgojo L, Van't Veer P, Fernández-Crehuet J, Kok FJ, Martin-Moreno JM. Toenail cerium levels and risk of a first acute myocardial infarction: the EURAMIC and heavy metals study. Chemosphere 2006;64(1): 112–20.

[49] Ali SH. Social and environmental impact of the rare earth industries. Resources 2014;3(1):123–34.

[50] Hirano S, Suzuki KT. Exposure, metabolism, and toxicity of rare earths and related compounds. Environ Health Perspect 1996;104:85–95.

[51] Takahashi Y, Kondo K, Miyaji A, Watanabe Y, Fan Q, Honma T, Tanaka K. Recovery and separation of rare Earth elements using salmon milt. PLoS One 2014;9(12):e114858.

[52] Royen H, Fortkamp U. Rare earth elements - purification, separation and recycling. Stockholm (Sweden): IVL Swedish Environmental Research Institute; 2016. 34 p. Report No.: C211.

[53] Jin FR, Oh JH. The analysis on the strategic protection of China's rare earth resources and its implications. Sejong (Korea): Korean Institute for International Economic Policy; 2011. 113 p. Report No.: 11-23.

[54] US EPA (United States Environmental Protection Agency). Risk assessment guidance for superfund volume I human health evaluation manual (Part A) [Internet]; 1989 [cited 2019 Mar 27]. Available from: https://www.epa.gov/sites/production/files/2015-09/documents/rags_a.pdf.

[55] Xu T, Peng HQ. Formation cause, composition analysis and comprehensive utilization of rare earth solid wastes. J Rare Earths 2009;27:1096–102.

[56] UNEP (United Nations Environment Programme). Recycling rates of metals: a status report [Internet]; 2011 [cited 2019 Mar 27]. Available from: http://www.resourcepanel.org/reports/recycling-rates-metals.

[57] Du X, Graedel TE. Global in-use stocks of the rare earth elements: a first estimate. Environ Sci Technol 2011;45:4096–101.

[58] Walton A, Williams A. Rare earth recovery. Mater World 2011;19:24–6.

[59] BLS (Bureau of Labor Statistics). National census of fatal occupational injuries in 2016 [Internet]; 2017 [cited 2019 Mar 27]. Available from: https://www.bls.gov/news.release/archives/cfoi_12192017.pdf.

[60] ICMM (International Council on Mining & Metals). Good practice guidance on health impact assessment [Internet]; 2010 [cited 2019 Mar 27]. Available from: https://www.icmm.com/en-gb/publications/health-and-safety/good-practice-guidance-on-health-impact-assessment.