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The EURARE Project: Development of a Sustainable Exploitation Scheme for Europe’s Rare Earth Ore Deposits

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Numerous European industries are heavily dependent on imported rare earth element (REE) raw materials. This has created a need for the European Union (EU) to ensure a sustainable supply of REE minerals, as well as develop from the ground up the currently non-existent European REE extraction and processing industry. In order to support this, the European Commission, though the Seventh Framework Programme (FP7) scheme, funded the EURARE project which runs from 1st January 2013 to 31st December 2017. Through the EURARE project, selected European REE deposits have been researched and in certain cases identified resources were successfully processed for REE production. Several REE deposits across Europe have been the focus of detailed geological field and laboratory work. Mineral concentrates obtained from the Norra Kärr deposit in Sweden, the Kringlerne deposit in Greenland and the Kvanefjeld deposit in Greenland, Rødberg ore from Norway and bauxite residue from Greece were tested from laboratory to pilot scale by means of conventional and innovative metallurgical processing. The novel technologies developed provide efficiency and selectivity in various steps of the metallurgical processing, from ore beneficiation to metal production. A road map for sustainable REE production in Europe is now being developed, which includes an evaluation of the environmental benefits and risks of the EURARE technologies.

1. Introduction

The REE are a group of 17 elements, comprising the elements scandium (Sc), yttrium (Y) and the 15 lanthanides (elements no. 57–71) as defined by the International Union of Pure and Applied Chemistry (IUPAC). The REE rank among the most critical of raw materials to Europe due to the risk of supply interruption from the major producer, China, and their significant economic importance (1).

REE are not currently exploited in Europe, but several REE projects are being explored or being technically or economically assessed currently, with some having reached an advanced stage of exploration and development (for example, pilot beneficiation and extraction studies to pre- or final feasibility studies). Examples include the alkaline igneous rock-hosted deposits Kvanefjeld and Kringlerne, in South Greenland, the Norra Kärr deposit in Sweden, and the heavy mineral ash fall or placer deposit, Aksu Diamas in Turkey. A number of carbonatite-hosted REE projects are also being explored, including Fen in Norway and Sarfartôq in Greenland, but have not yet reached an advanced stage of development.

Several REE occurrences and deposits across Europe have been identified in the course of the EURARE project, and an overview of the REE-occurrences and deposits in Europe has been compiled by the EURARE project and published (2). EURARE project partners have carried out research on several potential REE deposits in Europe. These include a number of deposits in the Nordic countries, such as the Basnäs skarn-hosted REE mineralisation (3) and REE enrichment in the magnetite-dominated ore of the Kiruna deposit (4) in Sweden. In Norway, research has included characterisation of the apatite resources and their potential for REE mineralisation (5) and the investigation of REE anomalies in the Nordkinn Peninsula (6). The potential for REE mineralisation in Greenland was highlighted in several papers and reports (7) while new discoveries of REE mineralisation in the Ilímaussaq Intrusion have also been identified (8, 9). Additional research has been undertaken in Turkey and Greece, where the potential for REE resources in bauxites and red muds has been investigated (10). The link between alkaline volcanism and the potential for REE deposits in related ash falls has also been explored (11).

EURARE has created an online, open access, database combining geographical, mineralogical and technological data, termed the Integrated Knowledge Management System (IKMS). At the time of publication the EURARE dataset in the IKMS contains data for 156 REE occurrences across Europe (**Figure 1**). The IKMS contains over 150 documents related to these occurrences and REE processing technologies.

The data gathered through the EURARE project highlights key issues such as the 'balance problem' between demand and natural abundance of the REE (12) and has been used to aid the development of a method for assessment of REE demand, which is of particular importance for European policy makers (13, 14).

2. European REE resources

The EURARE project studied some of both the most advanced and promising REE resources in Europe:

Norra Kärr: Leading Edge Materials (formed in August 2016 *via* the merger of Tasman Metals Ltd with Flinders Resources Ltd), is the license holder of Norra Kärr, located in south-central Sweden, reported a probable NI 43-101 compliant resource of 23.6 million tonnes, grading at 0.592% total rare earth oxide (TREO). The preliminary feasibility study suggests an annual mining operation of about 1.150 million tonnes per annum (tpa), equivalent to about 6800 tpa TREO, of which 3611 tpa are heavy rare earth oxides (HREO). The estimated lifetime is 20.5 years.

Kvanefjeld: Greenland Minerals and Energy Ltd (GME), the license holder of the Kvanefjeld REE-deposit, located in southern Greenland, reported a measured Joint Ore Reserves Committee (JORC) 2012 compliant resource of 143 million tonnes, grading at 1.2% TREO, 303 ppm U_3O_8, and 0.24% zinc (Zn), equivalent to about 1.72 million tonnes TREO. GME signed a memorandum

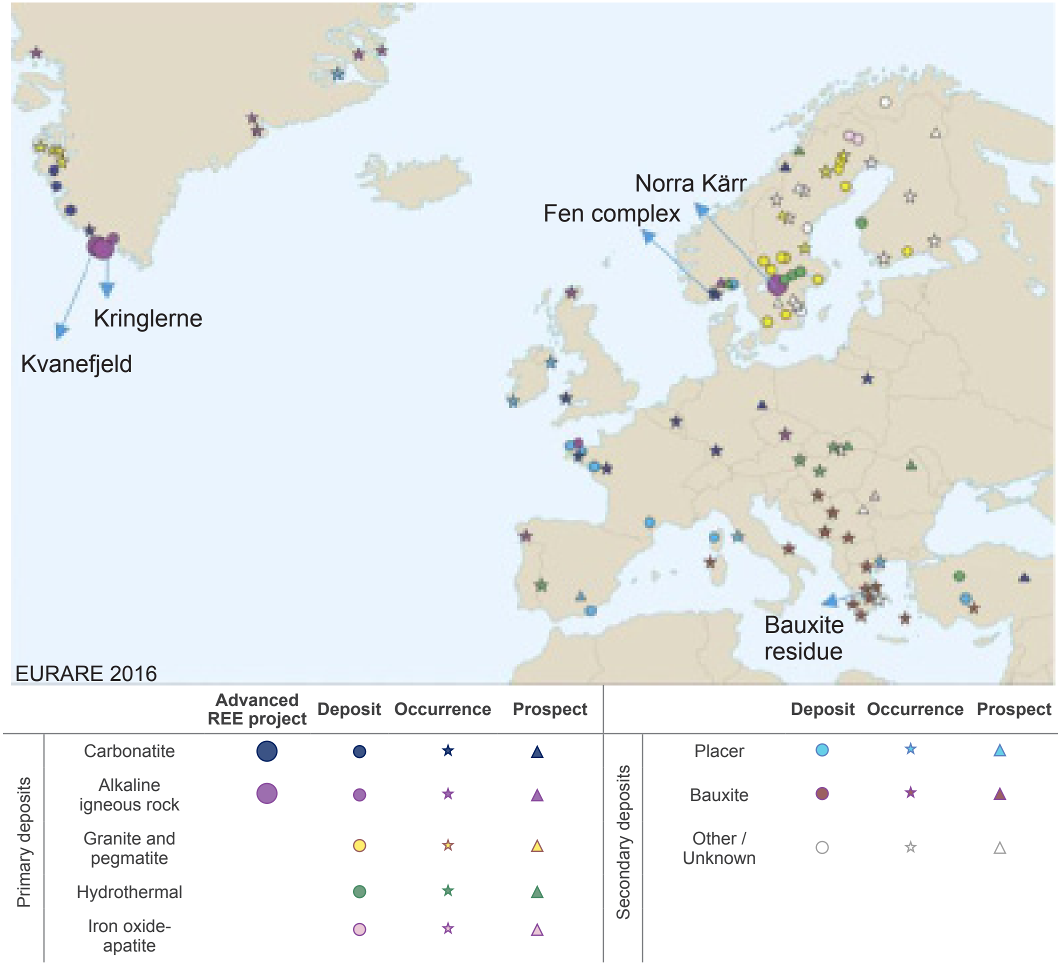

Fig. 1. European REE occurrences and deposits as listed in the EURARE-Integrated Knowledge Management System

of understanding with the Chinese REE group, Shenghe Resources Holding Co, aimed at technical and commercial collaborations. GME applied for the exploitation lease in 2015.

Kringlerne: Tanbreez Mining Greenland AS, the license holder of the Kringlerne project, located *ca*. 10 km south of Kvanefjeld, South Greenland, reports "the current inferred resource is more than 4.7 billion tonnes of eudialyte bearing ore, which contains variable contents of extractable rare earth". Tanbreez reports grades as follows: light rare earth elements (LREE): 0.5% and heavy rare earth elements (HREE): 0.15%. In addition they report the presence of valuable minerals such as 1.8% ZrO_2; 0.2% Nb_2O_5; and 0.02% Ta_2O_5. The detailed geological data are not disclosed. An estimated mine throughput of approximately 1 million tonnes per annum is envisaged to generate about 6500 tpa TREO. The mining license for the Kringlerne project is currently under application.

Fen: REE Minerals AS holds exploration rights on Fen carbonatite deposit in southern Norway. It is reported to contain 486 million tonnes, of søvite rock type only, grading about 0.9% TREO, and is therefore considered among the largest REE deposits in the world.

Aksu Diamas: This is a heavy mineral ash-fall or placer deposit in eastern Turkey. It contains a reported 495 million tonnes with a grade of 0.1% TREO. The REE would be produced as a byproduct of magnetite production from this deposit. Pilot plant production of the rare earth-bearing minerals using gravity and flotation methods were established; however, exploration and development of this project halted in summer 2014.

Other European projects that have been explored to the stage of informal resource estimation, but now been put on hold include: Sarfartôq (REE in pyrochlore) and Motzfeldt (REE in bastnäsite, columbite, eudialyte) in Greenland, Olserum (REE in fluorapatite, monazite, xenotime) in Sweden, and Storkwitz (REE in parasite, röntgenite, apatite) in Germany. The REE resources of each of the projects Kvanefjeld, Kringlerne and Norra Kärr alone could potentially secure European REE supply for decades to come (15).

Going to secondary REE resources, the bauxite residue from Greece, a metallurgical byproduct from alumina refining through the Bayer process, was found to contain about 0.14% of TREO. Taking into account the bauxite residue production per year it is estimated that a full REE exploitation just from Greece can contribute 6–12% of the European demand in REE such as neodymium (Nd), Y, cerium (Ce) and lanthanum (La) while simultaneously contributing to solve the bauxite residue landfill issue (16).

3. Metallurgical Exploitation of Selected European REE Ores

The industrial, academic and research and development (R&D) synergies for developing a European metallurgical exploitation based on REE primary and secondary resources can be seen in **Figure 2**.

3.1 Ore Beneficiation Studies

The beneficiation techniques of available European REE ores were optimised targeting the production of mineral concentrates with minimal environmental consequences at the Geological Survey of Finland (GTK), Finland, and the Greek Institute of Geology and Mineral Exploration (IGME), Greece. The following tasks were generally completed for each of the REE ores: chemical analysis; mineralogical characterisation; beneficiation testing by gravity, magnetic separations, flotation and hydrometallurgical leaching, followed by general beneficiation flowsheet research and amenability evaluations. Meanwhile, technologies which minimise the consumption of energy and materials, and the environmental impact were investigated with laboratory test work. European REE hosting minerals that were studied for metallurgical exploitation during this project are summarised in **Table I**.

The novel beneficiation technologies developed and optimised for the three European REE ores from Kvanefjeld, Kringlerne and Norra Kärr deposits were demonstrated by pilot testing. **Figure 3** shows the pilot beneficiation operation for the Kvanefjeld ore conducted at the pilot plant of GTK Mintec in Outokumpu, Finland.

3.2 Hydrometallurgical Studies

In Europe the majority of the REE deposits and resources host REE mineralisation in silicate minerals. In literature, hydrometallurgical studies of REE extraction from silicate minerals are scarce. The dissolution of eudialyte has been the focus of past research (17); however, further results of these studies are difficult to attain. Studies of metallurgical extraction of REE from the steenstrupine mineral at Kvanefjeld

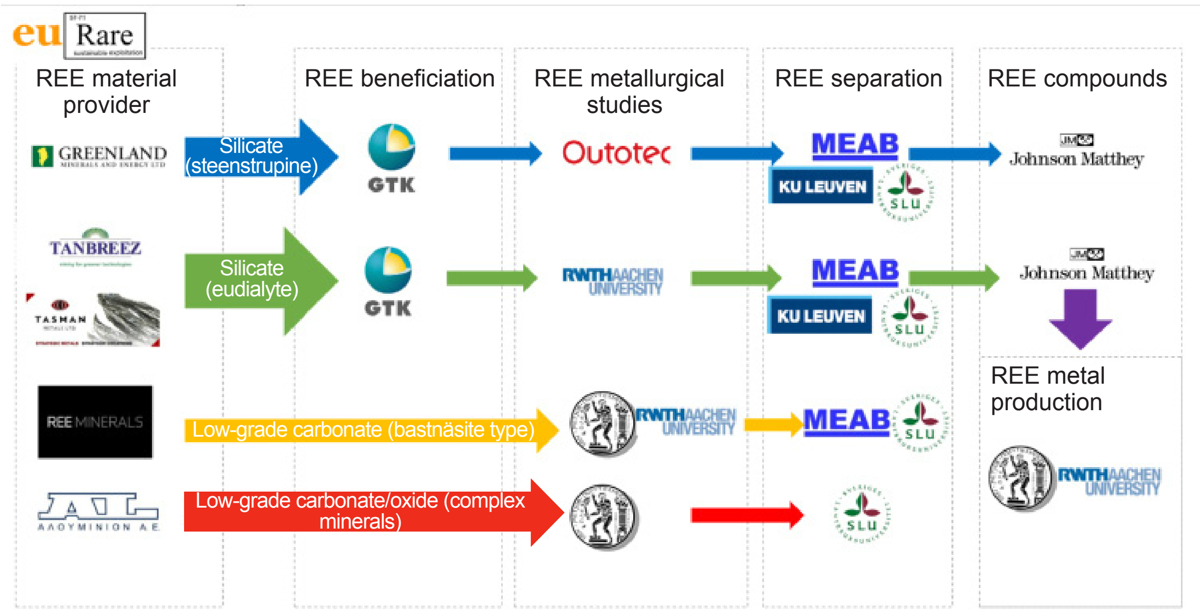

Fig. 2. Industrial, academic and R&D synergies for developing European metallurgical exploitation based on REE primary and secondary resources in the EURARE project

Table I Minerals Explored for Beneficiation in the EURARE Project

Provider	Scale of test	Ore conc.	%REO	REE host mineral	Type	Beneficiation
Greenland Minerals and Energy Ltd (GME)	Pilot	Kvanefjeld	14.15	Steenstrupine	Silicate	Froth flotation
TASMAN (Currently Leading Edge Materials)	Bench	Olserum	20	Monazite-xenotime	Phosphate	Froth flotation
TANBREEZ	Pilot	Tanbreez	2.03	Eudialyte	Silicate	Wet high intensity magnetic separation
TASMAN (Currently Leading Edge Materials)	Pilot	Norra Kärr	1.65	Eudialyte	Silicate	Wet high intensity magnetic separation
Greek Placer Deposit	Lab	Peramos	1.5	Allanite	Silicate	Dry magnetic separation
FEN (currently REE Minerals AS)	Lab	Rødberg	1.04	Bastnäsite	Carbonate	Insusceptible

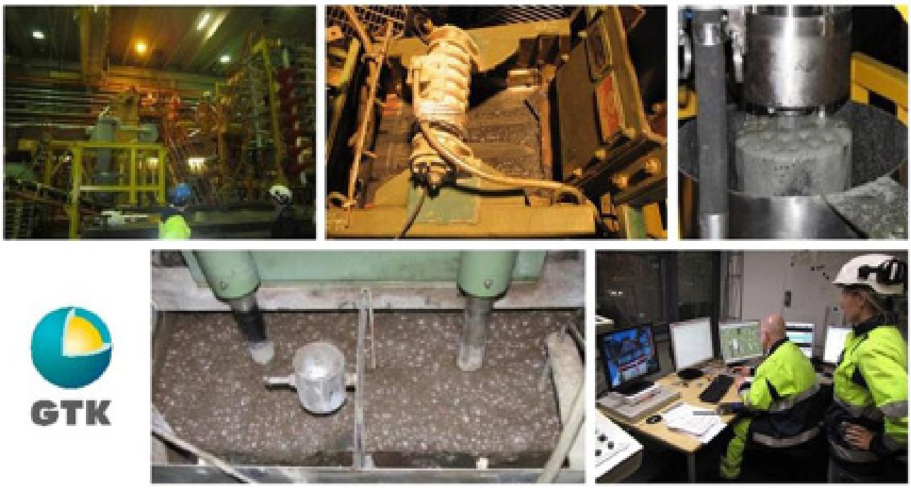

Fig. 3. Kvanefjeld ore froth flotation beneficiation pilot in GTK, Finland

also is not covered in the literature. The main issue upon dissolving REE silicate minerals is the formation of silica gel which hampers further solid liquid separation, resulting in an incomplete process.

During extensive research and experimentation in Rheinisch-Westfälische Technische Hochschule (RWTH) Aachen, Germany, and National Technical University of Athens (NTUA), Greece, it was found that the silica gel issue can be addressed by controlling the silica gel formation during leaching in various ways. Treatment of eudialyte concentrate was performed with hydrochloric acid in the demonstration plant at the Process Metallurgy and Metal Recycling (IME) laboratory of RWTH Aachen (**Figure 4**). The fuming process developed (18) represents the first use of this hydrometallurgical strategy in order to process rare earth elements from concentrate into solution whilst avoiding silica gel formation.

GME successfully completed the refinery pilot plant operations in Finland for the Kvanefjeld Project (**Figure 5**). Pilot runs conducted at Outotec Pori Research Laboratories, Finland, as part of the EURARE programme produced ~25 kg of mixed rare earth carbonate. This was used by EURARE partners to produce advanced rare earth products by separation techniques described further below.

Fig. 4. Treatment of eudialyte concentrate was performed in the demonstration pilot plant at the IME Process Metallurgy and Metal Recycling laboratory of RWTH Aachen, Germany

Direct acid leaching of Rødberg ore from Fen minerals (currently REE Minerals AS), successfully resulted in high REE extraction yields; however, the presence of acid consumable minerals such as calcite remains an issue. An innovative process developed by NTUA (19) based on the functionalised ionic liquid betainium bis(trifluoromethylsulfonyl)imide [Hbet][Tf$_2$N] (20) was also developed for leaching REE from Rødberg ore. The key aim of using ionic liquid solvents is to develop a process for low grade REE resources that results in the generation of a final aqueous REE-enriched pregnant solution for further purification.

[Hbet][Tf$_2$N] leaching was first applied to bauxite residue from Greece. The great innovation of [Hbet][Tf$_2$N] is its ability to selectively dissolve REE *versus* iron (Fe), silicon (Si) and titanium (Ti) from complex metal matrices. Additionally the metals dissolved in the ionic liquid can be extracted upon contact with an acidic solution producing a pre-concentrated REE solution for further purification. The ionic liquid subsequently regenerates and can be reused for the leaching step (19). The ionic liquid-based process was further validated at a bench scale of 4 l applied to both bauxite residue and Rødberg ore (**Figure 6**).

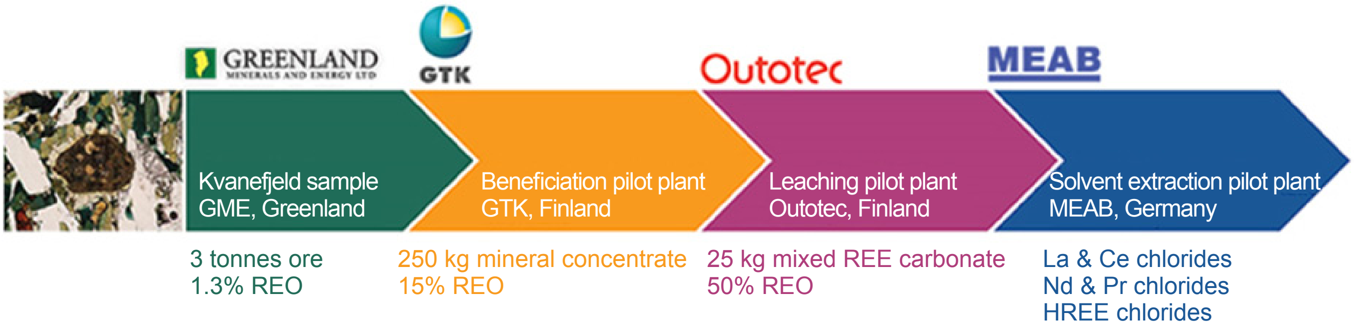

Fig. 5. Pilot achievements on the Kvanefjeld ore

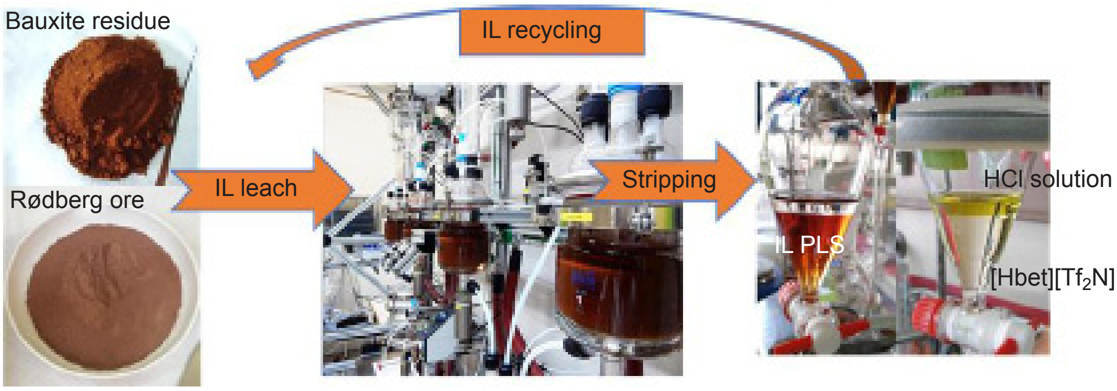

Fig. 6. Ionic liquid leaching of Rødberg ore and bauxite residue. IL = ionic liquid; PLS = pregnant leach solution. Visual flowsheet with demonstration prototype plant designed by MEAB and built in NTUA, Greece

3.3 REE Separation

The EURARE project developed procedures for the separation and extraction of REE from mixed carbonates, the result of the initial processing. MEAB Chemie Technik GmbH (MEAB), Germany; the Swedish University of Agricultural Sciences (Sveriges Lantbruksuniversitet, SLU), Sweden; and Katholieke Universiteit (KU) Leuven, Belgium, worked on three different approaches. MEAB worked on the separation of REE through conventional solvent extraction employing standard organic phases, SLU investigated the REE separation through innovative ion exchange, applying magnetic modified nano-adsorbents (21) whereas KU Leuven studied the REE separation through innovative solvent extraction using ionic liquids as both diluents and a source of coordinating anions (22). MEAB has successfully commissioned their solvent extraction demonstration plant in Aachen, Germany (**Figure 7**). Working with REE carbonate produced from hydrometallurgical studies has resulted in the separation of heavy (HREE: holmium-lutetium (Ho–Lu), Y), medium (MREE: samarium-dysprosium (Sm–Dy)) and light (LREE: La–Nd) rare earth elements. Y was separated from the HREE fraction and a mixture of praseodymium (Pr) and Nd was separated from the LREE fraction. Finally, Y was successfully separated from the LREE fraction (the raffinate arising after undergoing split-anion extraction). The main process uses bases for conditioning the organic phase and controlling the pH of the aqueous phase, acidic extractants for extracting the targeted REE into the organic phase, 0.5–1.5 M hydrochloric acid (HCl) for scrubbing unwanted REE from the organic phase at high organic:aqueous volume ratios (o:a) and 4 M HCl for stripping the required REE from the organic

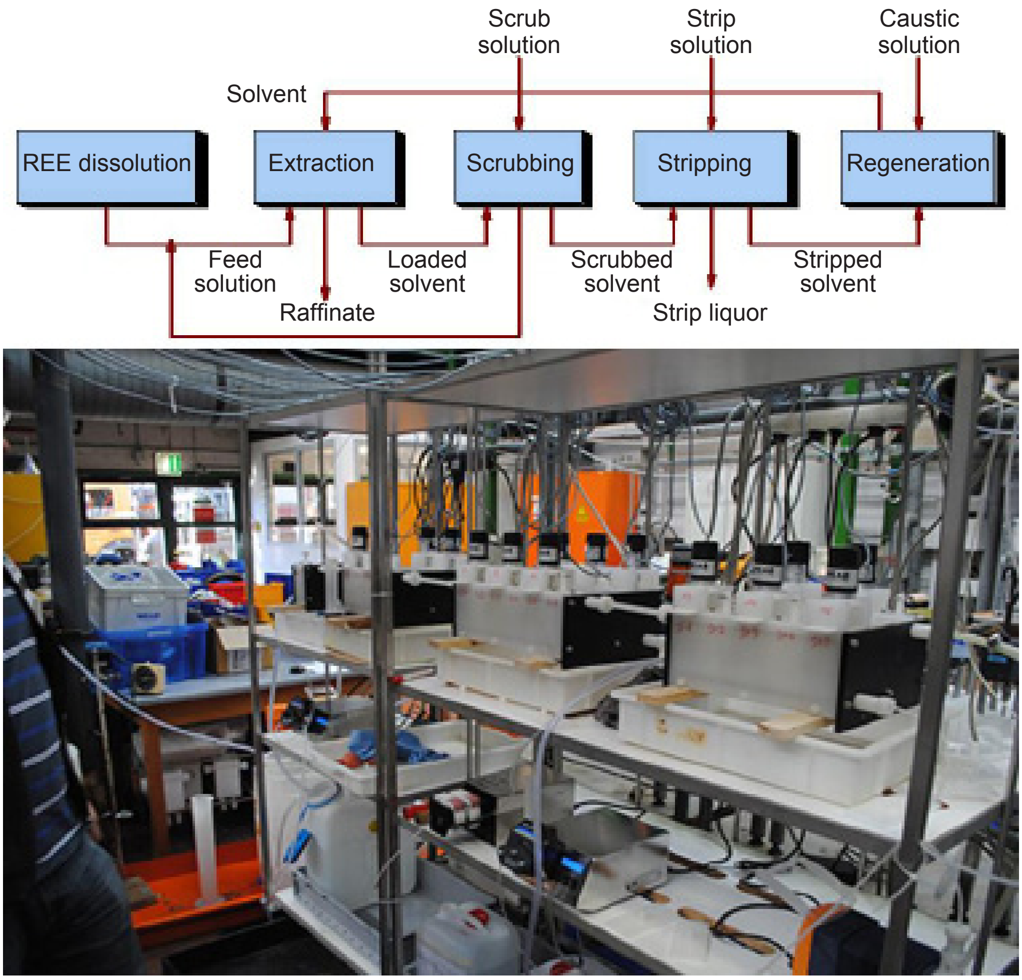

Fig. 7. General Process Flow Sheet – REE separation and purification by solvent extraction (MEAB, Germany). Demonstration pilot

phase. The process was tested by MEAB in continuous extraction demonstration tests using a multistage arrangement of mixer settler units (**Figure 7**).

Additionally the innovative ion exchange work performed by SLU has resulted in the preferential extraction and separation of Dy, Y, Nd and LREE. In one step, the sum of REE is first separately extracted from the other major metals, like Fe, zirconium, aluminium and calcium that are usually present as impurities in the pregnant solution. The next steps in the process exploit magnetic silica (SiO_2) based nanoparticles functionalised by different organic ligands for selective separation of individual REE. The nanoadsorbents are then easily separated from solution by magnet. Back extraction (stripping) of the wanted REE from the adsorbent is made by a controlled decrease in pH (**Figure 8**). For improving selectivity of the adsorbents in stripping and controlling the pH of the aqueous phase, 0.1 to 1 M of nitric acid (HNO_3) and 5 wt% of ammonia (NH_3) are employed, permitting the volumes of employed acids and bases to be strongly reduced. The process is green, employing only the aqueous media and enjoys recycling of the adsorbent phase.

Split-anion extraction is a new method of solvent extraction developed at KU Leuven for the separation of REE, without changing the aqueous phase. The extraction of REE from aqueous chloride feed solutions is carried out using a neutral extractant dissolved in an ionic liquid (diluent) which contains the REE-coordinating anions. The anion of the ionic liquid (NO_3^- or SCN^-) coordinates and transports the $REECl_3$ from the aqueous phase into the water-immiscible ionic liquid. The work performed by KU Leuven has resulted in the separation of HREE and MREE from LREE and Y from the chloride route without using acidic extractants. This created major improvements to the current solvent extraction technology with particular regard for cost, health, safety and environmental issues such as:

- The selective separation of REE into groups from chloride aqueous solutions requires less capital expenditure and an easier wastewater treatment than from nitrate aqueous solutions
- The replacement of organic solvents by non-fluorinated ionic liquids involves low volatility, non-flammability and no accumulation of static electricity
- Using neutral extractants means little to no need for pH control and easy stripping by water, and this is reflected in reduction in consumption of chemicals, lower operating costs and easier wastewater treatment (**Figure 9**).

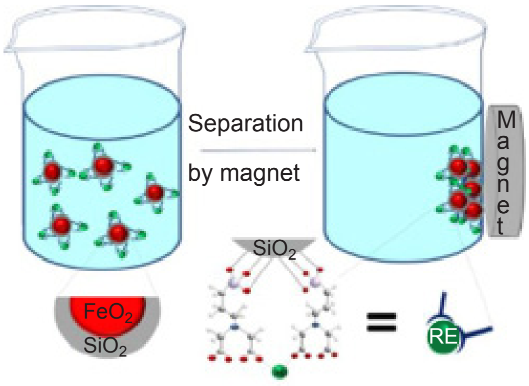

Fig. 8. Schematic presentation of hybrid magnetic adsorption of REE developed from SLU

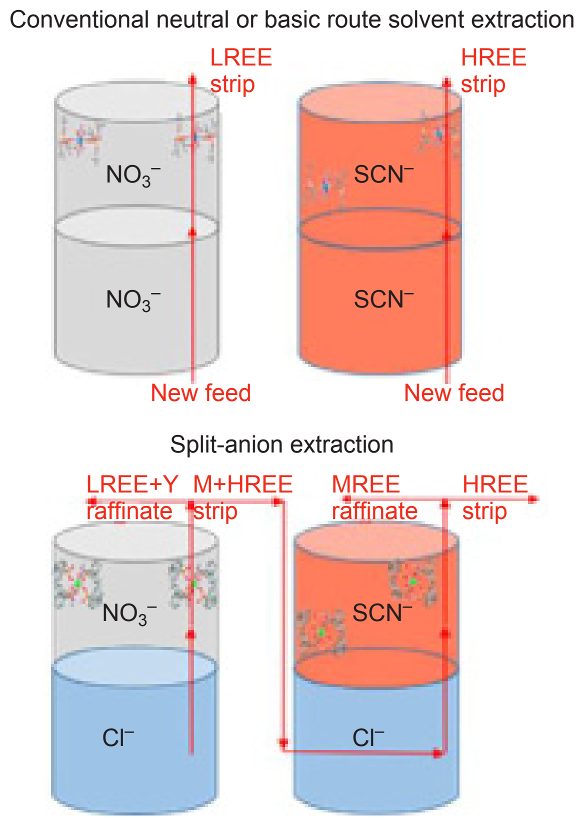

Fig. 9. General representation of the separation of REE by split-anion extraction developed by KU Leuven, Belgium. Comparison with conventional route

3.4 REE Metal Production

The challenges of applying REE molten-salt electrolysis under European environmental standards led RWTH to develop an automated new cell design (closed system) with the main goal of improving current molten salt technology. Key concerns are the reduction of energy losses, improved cost efficiency and reducing impurity interferences. The developed cell (**Figure 10**) resulted in process parameter optimisation, automation, reduction of greenhouse gasses evolution, and high purity metal production for Nd and didymium (Nd-Pr) metals.

During the EURARE project, the production of metallic rare earths by the use of novel electrolytes, such as the ionic liquids, was researched in NTUA. The main goal was to resolve a metallurgical process based on ionic liquids for the production of rare earths by avoiding the currently used technology of high-temperature molten salt electrolysis. In the first phase of the project, various kinds of ionic liquids were tested, in order to find the most suitable among them that would permit the reduction of rare earths prior to their decomposition. In the second phase, the most promising ionic liquids were investigated. More precisely, the reduction of rare earth cations La^{3+}, Sm^{3+}, Nd^{3+}, Dy^{3+} to the metallic state and their subsequent electrodeposition in the ionic liquids *N*-butyl-*N*-methylpyrrolidinium bistriflimide ([BMP][TF_2N]) and trimethyl butylammonium bistriflimide ([Me_3NBu][TF_2N]) were studied. These hydrophobic ionic liquids present a wide electrochemical window and a suitable ionic conductivity rendering them promising electrolytes for rare earths reduction. Scanning electron microscopy (SEM) and electron dispersive spectroscopy (EDS) analysis reveal that rare earths electrodeposition is feasible with the use of ionic liquids as electrolytic media (**Figure 11**).

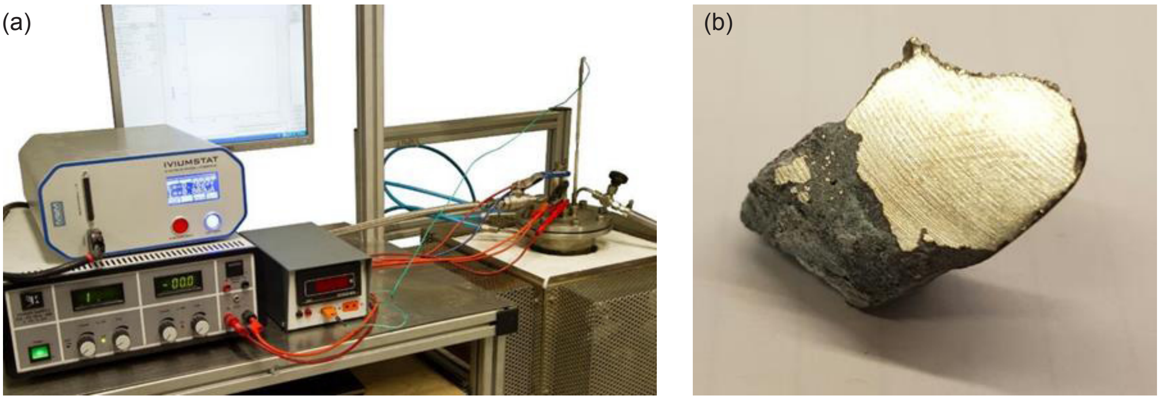

Fig. 10. (a) Optimised molten salt electrolysis cell setup; (b) didymium alloy obtained by electrodeposition in PrF_3-NdF_3-LiF electrolyte at IME Process Metallurgy and Metal Recycling laboratory of RWTH Aachen, Germany

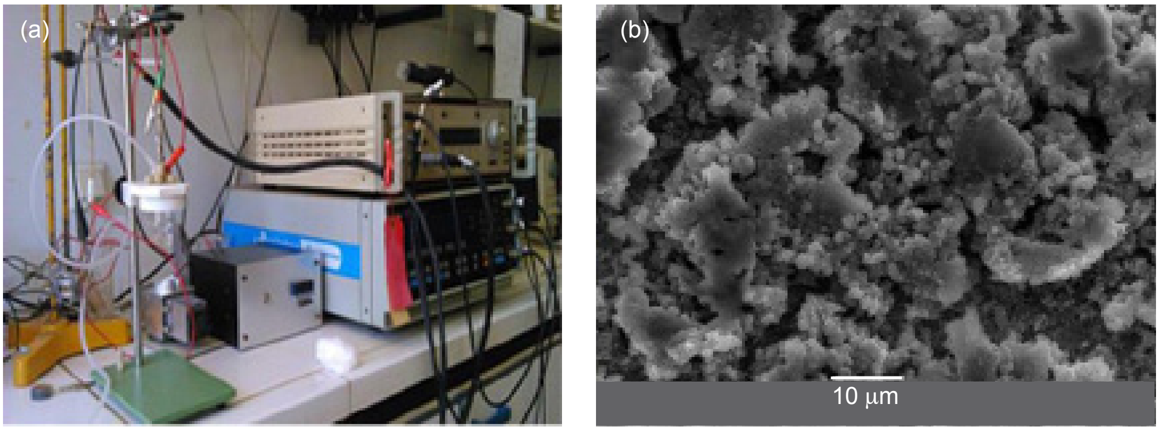

Fig. 11. (a) Cyclic voltammetry equipment; and (b) SEM image of Nd deposition at the Laboratory of Metallurgy, NTUA, Greece

3.5 Pilot Plants

The developed technologies in REE metallurgical extraction from European resources are further being evaluated on a pilot scale starting with 1–5 tonnes of ore concentrate to produce 5–50 kg of REE. The demonstration pilot plant starting with 3 tonnes of Kvanefjeld ore has been successfully concluded (**Figure 5**) producing separate streams of REE chloride groups, whereas the Norra Kärr and Kringlerne pilot plant demonstrations are currently under development.

4. Roadmap for Sustainable REE Exploitation and REE Material Supply Autonomy in Europe

The overall aim of the EURARE project is to develop a sustainable exploitation scheme, or roadmap, for Europe's REE ore deposits that will result in REE supply autonomy in Europe. This requires an economically viable REE ore resource, effective beneficiation, hydrometallurgy and separation techniques, and efficient metal production techniques. However, it also means that the best available techniques should be applied with regard to emissions and impacts on the environment as a whole.

Activities in the EURARE project relating to sustainability and environmental impact assessment will therefore contribute to the roadmap. The European legislation in place to limit the environmental impacts of the REE industry has been reviewed and compared with international best practice (23). Life cycle assessment is being used to quantify the energy and resources used in the processes and the waste, emissions and greenhouse gases produced per unit mass product. This will be supported by a compilation of the hazards associated with the chemicals used. The tailings, tailings water and process water generated during the beneficiation of selected EURARE ores have been characterised, and wastes from the EURARE hydrometallurgical and separation processes are now being characterised. These characterisations allow the wastes to be defined in terms of European legislation and therefore identify the waste management options available. The data from the beneficiation processes will also be used to evaluate stabilisation methods for the tailings and wastewater treatment technologies to allow the reuse or discharge of wastewater and determine whether the tailings are suitable for use in the construction industry.

5. Summary of Achievements

The establishment of a critical mass of scientists and engineers to support the REE exploitation, processing and manufacturing industry has resulted in great advancements in the European REE exploitation sector.

- ✓ Several occurrences and advanced projects of REE resources have been explored in Europe. EURARE has created an online open access database combining geographical, mineralogical and technological data, the IKMS. At the time of this publication the EURARE dataset in IKMS contains 156 REE occurrences across Europe. The IKMS contains over 150 documents related to these occurrences and REE processing technologies.
- ✓ The REE resources for each of the projects Kvanefjeld, Kringlerne, Norra Kärr, Aksu Diamas and Fen complex could potentially secure European REE supply for decades to come.
- ✓ Conventional and innovative metallurgical treatment of European REE resources, starting from beneficiation, leaching and REE separation to metal production, were developed successfully.
- ✓ Under EURARE, pilot testing 'from ore to metal' is taking place, demonstrating the potential to develop a European-based REE extractive industry.
- ✓ Under the EURARE project the first European Rare Earth Resources Conference, ERES2014, was organised in September 2014, and its follow up, ERES2017 has been announced for May 2017.
- ✓ A roadmap is being developed for sustainable REE exploitation in Europe based on the resources identified, technologies developed and the environmental benefits and risks associated with the technologies.

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