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Primary and secondary sources of rare earths in the EU-28: results of the ASTER project

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Abstract

The ASTER project aims at establishing flows and stocks of certain critical rare earth elements at the scale of the EU-28, considering both primary and secondary sources. Material flow analyses were performed taking into account processes along the value chain (separation, manufacture, use, waste management) and including also lithospheric (geologic) stocks. While this paper focuses on fluorescent lamp phosphors (Tb, Eu, Y), a paper by Planchon et al. (this conference) addresses permanent magnets and batteries (Nd, Pr, Dy). Results suggest that given the magnitude of flows in the EU-28, the development of a mining project in Sweden and/or Greenland would contribute significantly to reducing heavy rare earth; e.g. Tb, criticality.

Introduction

Member state and European policies with respect to the safe supply of mineral raw materials aim at improving eco-efficiency¹. The objective of eco-efficiency is to increase the quantity of services per unit mass of raw material, while decreasing environmental impacts. One of the pillars of eco-efficiency is recycling. In order to efficiently position the development of innovative recycling processes, it is important to have a clear picture of flows of stocks of mineral raw materials all along the value chain. The on-going ASTER project, started in 2012 for a period of three years, aims at applying material flow analysis (MFA) to selected rare earth elements (REEs) for the EU-28. Certain rare earth elements are deemed “critical”, as they combine importance for strategic sectors of the economy with risks of supply shortage^{3, 4}. The recent JRC report² identifies six critical REEs (Dy, Eu, Tb, Y, Pr and Nd) that are precisely the REEs that were selected for the ASTER project.

Given the importance of recycling, the ASTER project combines an “application” approach with an “element” approach. Table 1 shows the correspondence between the two. These three applications were selected because of their importance in

terms of use of the selected REEs, and also because recycling processes for these applications either already exist or else are currently under development⁵.

Table 1: Correspondence between applications and REEs addressed in the ASTER project

| Application | REEs |
|--------------------|-------------|
| Phosphor powders | Eu, Tb, U |
| Permanent magnets | Nd, Pr, Dy |
| NiMH batteries | Nd |

While REEs such as Nd are used in a variety of other applications, it was considered of lesser importance to examine, e.g., flows of Nd in ceramics, as there are currently no realistic prospects of recycling Nd contained in these products. In this paper, we adopt the following distinction between light (LREE) and heavy (HREE) rare earth elements⁶:

- LREE : La, Ce, Pr, Nd, Sm ;
- HREE : Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu (plus Y).

Methods

A systemic approach

For the case of fluorescent light phosphors, the system that was analysed is described schematically in Figure 1. The figure illustrates a certain number of processes with, starting upstream in the value-chain: separation (S) of rare earth concentrates, fabrication (F) of trichromatic phosphor powders, manufacture (M) of fluorescent lamps (LFL and CFL; resp., linear and compact fluorescent lamps), use (U) of these lamps in the economy, waste management (WM) of end-of-life products and final elimination (landfill and/or dissipation in the environment). These various processes are linked by arrows that represent flows from one to another. Differences between flows into or out of a process imply an addition or subtraction to stock within the process. The dashed line in Figure 1 represents the limits of the studied system (the EU-28). However, an additional “process” appears in Figure 1 indicated as “L” (lithosphere). The lithosphere represents geological potentialities in the form of potential REE resources (as opposed to reserves⁷). For geological and economic reasons, the limits of this process were taken as continental Europe (including the Scandinavian shield and the Kola Peninsula) plus Greenland. Therefore this process straddles the limits of the EU-28.

It is important to note that the system analysed in this study is an “open” system: there are inflows and outflows to and from the system all along the value chain. This is in contrast with most previous REE MFA studies which have addressed global flows and hence a “closed” system⁸. In the latter case, flows can be “constrained” by world mining production, as reported by, e.g., the USGS.

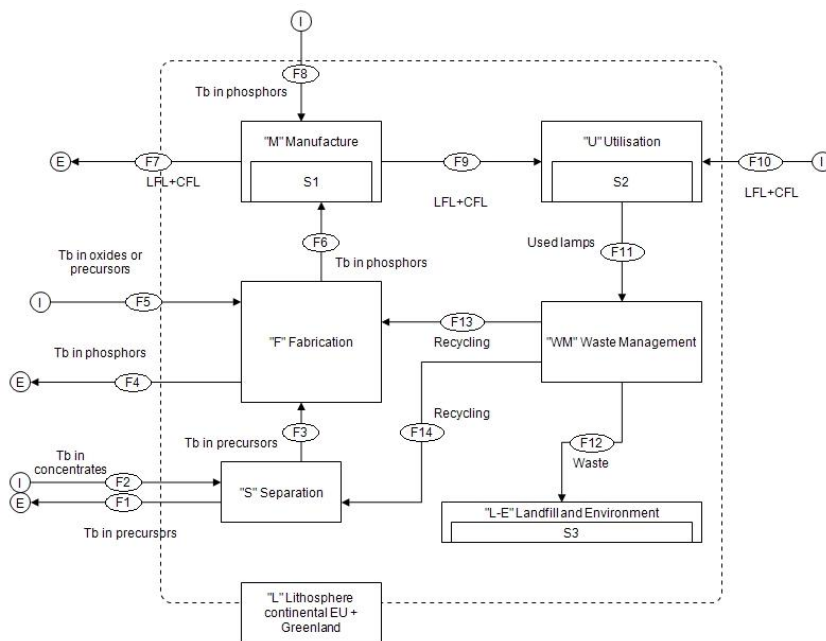


Figure 1: Schematic of the system investigated in the case of fluorescent-lamp trichromatic phosphors. LFL = linear fluorescent lamps; CFL = compact fluorescent lamps

Flows were estimated over a number of years exceeding the expected average lifetime of the considered products (taken as 6 years for fluorescent lamps), in order to obtain estimates of in-use stocks. Year 2010 was selected as the reference year for the final results of the calculations.

Information sources

Data mining constitutes the main task of any MFA. Sources of information included statistical custom data (EUROSTAT, Global Trade Atlas, ...), specialized reports^{10, 11}, USGS data¹², company reports on product sales, etc. With respect to the upstream portion of the value chain, the same HS codes (listed in Table 2) were used as in the Öko-Institut report¹¹. GTA data for Chinese exports based on 8-digit HS codes that are specific to individual REEs were also used.

These HS codes represent raw products that contain mixtures of REEs. In order to obtain data on individual REEs, a disaggregation method was applied¹³. The method

considers the relative market shares of products containing REEs (e.g., auto catalysts, magnets, batteries, polishing powders, etc.) and the composition of the REE mixtures in each product.

Table 2: HS codes used for REE-containing products in the upstream portion of the studied system

| HS code | Description in EUROSTAT |
|----------|--|
| 28053010 | Intermixtures or interalloys of rare-earth metals, Scandium and Yttrium |
| 28053090 | Rare-earth metals, Scandium and Yttrium (Excl. intermixtures or interalloys) |
| 28461000 | Cerium compounds |
| 28469000 | Compounds, inorganic or organic, of rare-earth metals, of Yttrium or of Scandium or of mixtures of these metals (Excl. Cerium) |

Certain data from EUROSTAT had to be corrected to account for the fact that Austria no longer reports its REE-related data after 2008. GTA data for Austria were disaggregated into imports and exports from intra- and extra EU-28, considering averages calculated over the pre-2008 period.

For the evaluation of geological potentialities in the lithosphere, over 350 deposits or occurrences located within the EU-28 were analysed¹⁴. The mineralogical characteristics of these deposits and occurrences provide information regarding potential resources, the reliability of which depends largely on the state of progress of exploration projects. However, the objective was to provide orders of magnitude for potential resources (lithospheric stocks), that could be compared with the magnitude of flows in the EU economy.

Uncertainties and reconciliation

Information collected in the course of a MFA for critical metals is inevitably fraught with uncertainty. This uncertainty is typically of an “epistemic” (reflecting incomplete knowledge) rather than a “stochastic” (reflecting random variability) nature. Uncertainty implies that MFAs do not balance and therefore reconciliation methods are used¹⁵. Such methods generally assume that uncertainty can be represented in the form of Gaussian probability distributions (see, e.g., the STAN software⁹).

However, epistemic uncertainty (such as typically ensues from expert-opinion) is more faithfully represented by the nested intervals (or fuzzy sets) of possibility theory¹⁶. Therefore, as part of the ASTER project, a methodology was developed for reconciling MFAs under fuzzy constraints^{17, 18}. This methodology was presented in 2014 at a workshop on data reconciliation in MFA, the proceedings of which can be obtained upon request (e-mail on 1st page of this paper).

Some results to-date

Results presented herein are preliminary as the project is still on-going. An important deliverable of the project will be Sankey diagrams for the considered REEs. These diagrams are described qualitatively below for the case of REEs in fluorescent light phosphors.

A first step of the disaggregation procedure consisted in converting tons of products corresponding to the customs data for the HS codes of Table 2, into tons of rare earth oxide mixtures (REOs). The conversion factors were selected based on data from USGS¹². For the metals and alloys (codes 280530), the conversion factor is 1.2 based on stoichiometric considerations. For codes 284610 and 284690, conversion factors were taken as, resp., 0.67 and 0.72.

Application of these conversion factors to historical imports and exports of raw products containing REEs in the EU-28 yields Figure 2. The significant decrease in imports observed in 2009 is interpreted as a consequence of the 2008 financial crisis.

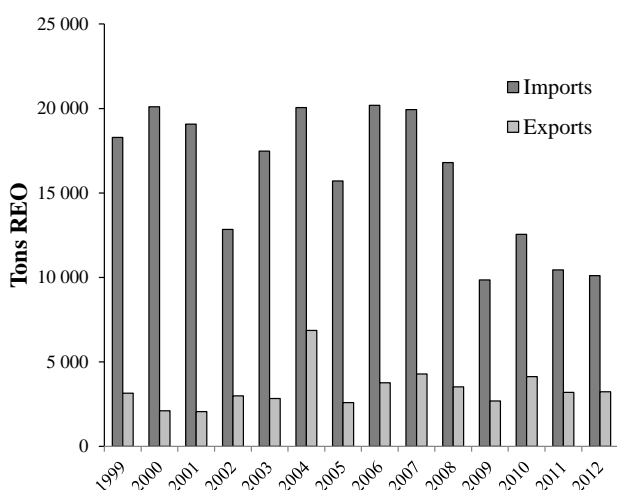


Figure 1: EU-28 imports and exports of REOs based on EUROSTAT (and GTA for Austria post-2008 correction)

Application of the disaggregation methodology to these data provides information relative to flows F1 and F2 of Figure 1, which can then be compared with information provided by SOLVAY, the only company in the EU-28 to perform separation of REEs for trichromatic phosphors. Separation is also performed in Estonia by SILMET, but not for phosphor applications (mainly for Nd). Further to separation, SOLVAY produces phosphor precursors that are sent to process “F” (Figure 1) to produce

phosphors. Information relative to imports and exports of phosphors is obtained based on HS code 320650 (*inorganic products of a kind used as luminophores, whether or not chemically defined*). Expert information was necessary to define the typical composition of these phosphors, taking into account that the composition is different for imported versus exported phosphors (in and out of the EU).

Further down the value chain, data was collected for manufactured products containing trichromatic phosphors. The two HS codes of Table 3 were used.

Table 3: HS codes for fluorescent lamps

| HS code | Description in EUROSTAT |
|----------|--|
| 31501530 | Fluorescent hot cathode discharge lamps (excluding ultraviolet lamps, with double ended cap) |
| 31501510 | Fluorescent hot cathode discharge lamps, with double ended cap (excluding ultraviolet lamps) |

Results of the analysis suggest that the orders of magnitude of Tb, Eu and Y flows in the EU-28 related to fluorescent lamp phosphors are resp., 10, 15 and 180 tons. Taking into account the history of flows into use in the EU-28 over a period exceeding the expected average lifetime of the fluorescent lamps, in-use stocks for Tb, Eu and Y in 2010 are estimated as, resp., 140, 220 and 2300 tons. The annual additions to in-use stock in 2010 are estimated to be resp., +8, +13 and +140 tons/yr for these elements.

With respect to flows F13 and F14 in Figure 1 (recycling), there was no recycling in 2010 in the EU-28, as SOLVAY started its recycling operations (in Saint-Fons and La Rochelle, France) in 2012. As an indication, the order of magnitude of Tb recycled each year is a few tons per year.

These figures can be compared to geological potentialities derived based on the analysis of three important REE projects: the Norra Kärr project in Sweden and two exploration projects in Greenland (Kvanefjeld and Kringlerne). The Norra Kärr project is well advanced and has recently received the authorization to operate from the Swedish government. We estimate the resource of Tb at this site to be on the order of 2 000 tons. The potentials of Kvanefjeld and especially Kringlerne are far superior (resp. 10 000 and 110 000 tons Tb), however, the likelihood of these projects entering into production is much lower, due in particular to environmental constraints and social acceptability. Nevertheless, these figures suggest that if just one of these projects enters the production phase (with Norra Kärr as the most likely candidate), the criticality with respect to certain heavy REEs such as Tb will be significantly reduced.

Conclusions

Results of the on-going ASTER project provide estimates of flows and stocks of certain REEs considered as critical. This paper addresses the results relative to REEs in fluorescent lamp phosphors, while another paper (Planchon et al., this conference) addresses Nd and Dy.

The data collected to-date suggest a somewhat “reassuring” picture with respect to criticality of Tb, Eu and Y. It is the opinion of the authors of this paper that the crisis regarding REEs that was experienced in 2011 is essentially over. It is reminded that in July 2011, peak prices of Dy and Tb exceeded average prices over the period 2002-2009 by factors on the order of, resp., 20 and 10, as a result of stricter quotas on Chinese exports. Several factors have contributed to reducing the pressure on these substances. On the one hand, countries that import REEs have diversified their sources of supply of LREEs (e.g., Mount Weld in Australia and Mountain Pass in the U.S.). Also, there are currently considerable efforts devoted to optimizing the quantities of REEs used in products, as for example Dy in permanent magnets. Substitution is also an important issue. Regarding Tb, the development of LED lighting, which use very little REEs and virtually no Tb, will contribute in the future to reducing the pressure on this element.

Detailed results of the ASTER project are currently in the process of being published. It is anticipated that the approach of associating both primary and secondary sources in the systemic analysis of critical metals will be applied to other substances (e.g., In, Ge, Ga, ...), as is the case for projects that have already been initiated. It should be noted however that, as for REEs, data regarding such “small” metals are hard to come by. On the one hand, there is very little information available regarding reliable estimates of resources⁶ for substances that are essentially by-products of carrier metals¹⁹. Also, considerable effort is still needed to quantify the quantities of these substances in products, where they may be present in very low concentrations (on the order of a ppm). Such information is crucial in order to evaluate the economic feasibility of recycling processes.

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References

1. COM. Roadmap to a resource efficient Europe. COM 571, European Commission, Brussels, 2011.
2. Moss, R., Tzimas, E., Willis, P., Arendorf, J., Thompson, P., Chapman, A., Morley, N., Sims, E., Bryson, R., Peason, J., Tercero-Espinoza, L., Sartorius, C., Ostertag, K. Critical metals in the path

- towards the decarbonisation of the EU energy sector. Assessing rare metals as supply-chain bottlenecks in low-carbon energy technologies. JRC Report EUR 25994 EN. 242 pp., 2013.
3. NRC. Minerals, Critical Minerals, and the U.S. Economy. U.S. National Research Council , Washington, D.C., National Academies Press, 245pp., 2008.
 4. EC. Critical raw materials for the EU. Report of the ad-hoc Working Group on defining critical raw materials. European Commission, June 2010.
 5. Binnemans, K., Jones, P.T., Blanpain, B., Van Gerven, T., Yang, Y., Walton, A., Buchert, M. Recycling of rare earths: a critical review. *Journal of Cleaner Production*, 51, 1-22., 2013.
 6. Wall, F. Rare earth elements. In: "Critical Metals Handbook", 1st edition (Ed. Gus Gunn), John Wiley & Sons, Ltd., 2014
 7. PERC. Pan-European standard for reporting of exploration results, mineral resources and reserves. Geological Service of Belgium, Brussels, Belgium, 2013.
 8. Du, X., Graedel, T.E. Uncovering the global life cycles of the rare earth elements. *Scientific Reports*, 1(145): 1-4, 2011.
 9. Brunner, P.H., Rechberger, H. *Practical Handbook of Material Flow Analysis*. Lewis Publishers, 2004.
 10. ROSKILL. *Rare Earths & Yttrium: Market Outlook to 2015*. 14th edition. Roskill Information Services Ltd, London, UK, 2011.
 11. Schüler, D., Buchert, M., Liu, R., Dittrich, S., Merz, C. Study on rare earths and their recycling. Öko-Institut Report, Darmstadt, Germany, 140 pp., 2011.
 12. USGS. *Mineral commodity summaries 2013*. U.S. Geological Survey, 198 pp., 2013.
 13. Goonan, T.G. Rare earth elements – End use and recyclability. U.S. Geological Survey Scientific Investigations Report 2011-5094, 2011.
 14. Charles, N., Tuduri, J., Guyonnet, D., Melleton, J., Pourret, O. Rare earth elements in Europe and Greenland: a geological potential? An overview. 12th meeting of the Society of Geology Applied to Mineral Deposits (SGA), 12-15 August 2013, Uppsala, Sweden, 2013.
 15. Kelly, J.D. Techniques for solving industrial nonlinear data reconciliation problems. *Computers and Chemical Engineering*, vol. 28, pp. 2837-2843, 2004.
 16. Dubois, D., Prade H. *Possibility Theory: An Approach to Computerized Processing of Uncertainty*, Plenum Press, New York, 1988.
 17. Dubois, D., Fargier, H., Ababou, M., Guyonnet, D. A fuzzy constraint-based approach to data reconciliation in material flow analysis. *International Journal of General Systems* (in press), 2014.
 18. Dubois, D., Fargier, H., Guyonnet, D. Data reconciliation under fuzzy constraints in material flow analysis. EUSFLAT-2013. Eighth conference of the European Society for Fuzzy Logic and Technology, September 11-13, Milano, Italy, 2013.
 19. Verhoef, E.V., Dijkema, G.P., Reuter, M.A. Process knowledge, system dynamics, and metal ecology. *Journal of Industrial Ecology*, Vol 8, 1-2, pp. 23-43, 2004.